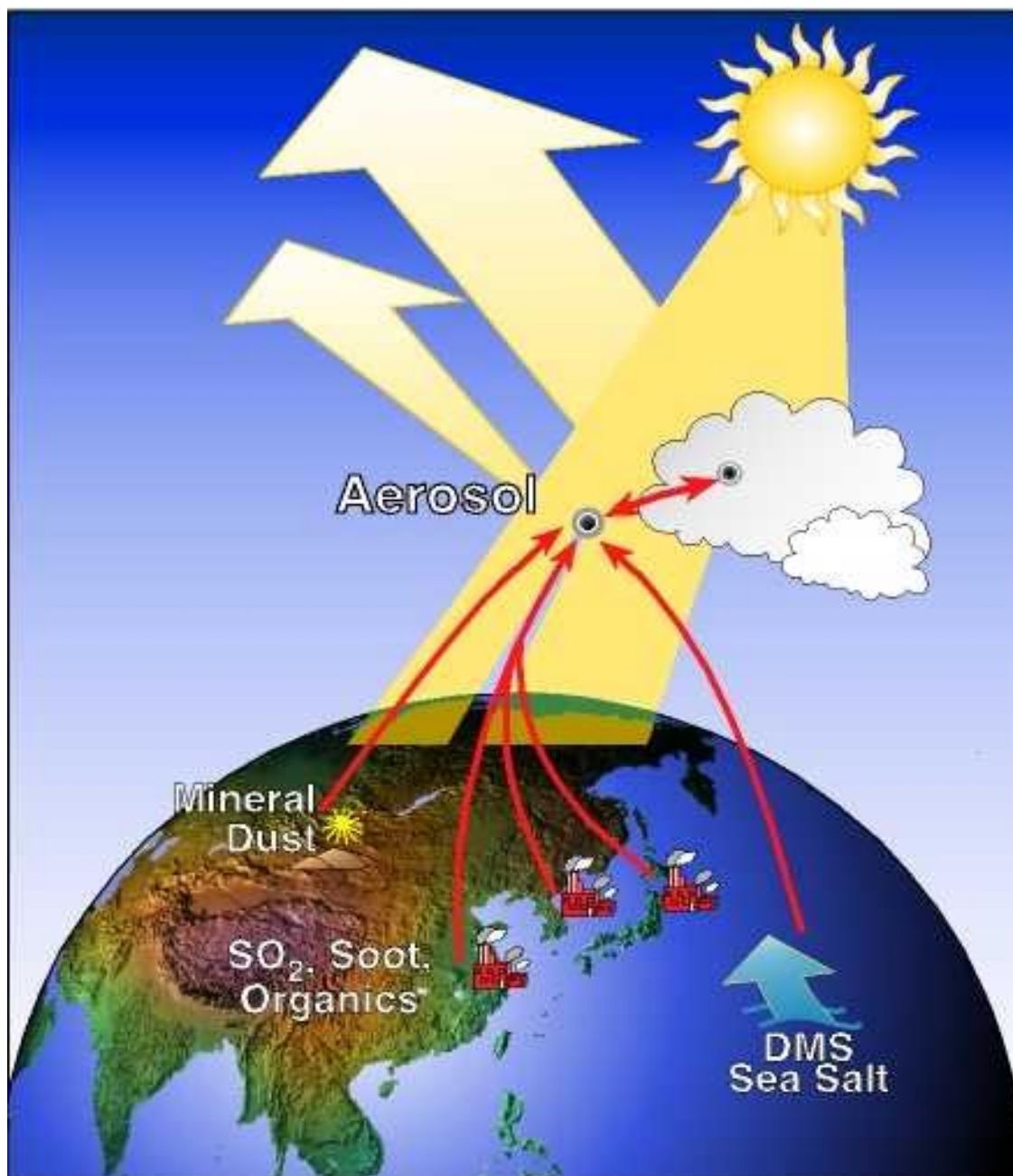


ACE-Asia



Survey and Evolution Component (AA-SEC)
NSF Large Field Program Scientific Overview

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ACE-Asia

Asian Pacific Regional Aerosol Characterization Experiment

Radiative Forcing due to Anthropogenic Aerosols Over the Asian Pacific Region

**Organized by the
International Global Atmospheric Chemistry Program's (IGAC)
Aerosol Characterization and Process Studies (ACAPS)
Asian and Pacific Regional Experiment (APARE) and
Marine Aerosol and Gas Exchange (MAGE) Activities**

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Further information on ACE-Asia can be found at:

<http://saga.pmel.noaa.gov/aceasia/>

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I. Executive Summary

The radiative forcing of climate by aerosols is one of the most significant environmental issues we face at the close of the 20th century. Governments are debating expensive measures to reduce greenhouse gas emissions, yet we cannot reliably model the climatic effects of different emission scenarios. One major reason is aerosols: the uncertainty in estimates of aerosol radiative forcing is the largest of any of the terms in the Earth's energy budget (IPCC, 1995). Even the sign is uncertain for both direct (scattering) and indirect (cloud-related) forcing. The major goal of ACE-Asia, an International Global Atmospheric Chemistry Program (IGAC) aerosol characterization experiment, is to make aerosol radiative forcing calculations more reliable, so that policymakers will have a better scientific foundation for the difficult mitigation decisions they will be forced to make in the next decade.

There are compelling reasons to study aerosols coming off eastern Asia into the western Pacific:

- A. Perhaps the most obvious one has just been stated: the remaining uncertainties (in models of aerosol impacts on climate) are still far too large to support informed decisions about ways to minimize future anthropogenically-caused climate changes. Our models need to be much more realistic.
- B. Thanks to recent developments, we are now able to measure aerosol concentrations and properties with an accuracy that will challenge models much more effectively.
- C. Asian anthropogenic emissions and mineral dust are very different from the environments of previous ACE experiments.
- D. Finally, expected increases in Asian emissions have the potential to cause large changes in radiation budgets, cloud microphysics, and hydrological output over the coming decades.

The objectives of the first ACE-Asia intensive experiment are to:

1. Survey the air leaving Eastern Asia to characterize aerosol physical, chemical, and optical properties and their variation with time and space, emphasizing variations with altitude and distance from shore;
2. Measure the evolution of aerosol physical and chemical properties in a Lagrangian framework, to identify controlling processes and infer their rates; and
3. Quantify the impact that aerosols have on the chemistry of the troposphere.

These studies will focus on four principal components of the aerosol: sulfates and other ionic species from urban/industrial emissions, mineral dust, carbonaceous material (black carbon, BC, and organic species), and sea salt. Although it is clear that sulfates are formed from SO₂, four different oxidation pathways (each with different product sizes) may control the conversion rate under various circumstances. Mineral aerosol is of concern both because it absorbs radiation (and thus can have a warming effect) and because it may serve as the core of a complex multicomponent aerosol (MCA) that can impact the transport and deposition of other species. BC also absorbs radiation, and may catalyze a variety of reactions on its surface. The quantity and identity of organics in aerosols has long been a mystery due to a lack of specific analytical techniques. Organics may substantially alter the response of aerosols to humidity changes (from our mostly-sulfate picture of just a few years ago): we now know that they may be as concentrated as sulfate in some places. Sea salt particles are emerging as potent chemical reactors, in which SO₂ can be oxidized (and rapidly removed), while they release halogen oxidants into the gas phase that may rival the importance of OH radical.

We propose to study Asian aerosols using three US mobile platforms (the NCAR C-130, the CIRPAS Twin Otter, and a NOAA or UNOLS ship) and 1-2 enhanced ground stations working with ships, aircraft, lidars, and surface sites from a variety of nations. Japan, Korea, China, and Taiwan have all formed national ACE-Asia committees and have committed funds for platforms and PIs. This multinational experiment will take place in March and April of 2001, with an operations base probably located in southern Japan.

Numerical models are the vehicles through which all our knowledge of aerosol sources, processing, removal, and effects will be codified. Our observations are being designed with the help of modelers to ensure that they are effective for testing and improving process, regional, and hemispheric aerosol models. We will make use of satellites both for experimental planning (such as locating dust layers) and for extrapolating results over larger scales. About half our resources will be devoted to surveying concentrations and properties vs. altitude and location, to generate spatially-gridded data in a variety of circumstances to challenge regional models. The other half will be devoted to Lagrangian experiments, in which we make repeated flights into a tagged airmass for 1-2 days, to improve process models, identify reaction pathways, and quantify rates. We hope to coordinate some of our flights with the NASA TRACE-P program, which will be focusing on photochemistry in Asian outflow at about the same time. If possible we will coordinate some observations with an airborne simulator for the PICASSO-CENA

spaceborne lidar.

The impact of Asian aerosols cannot be determined by extrapolation from experiments in other regions. Observations of properties and processes are needed in Asia and the northwestern Pacific to support the development and testing of realistic predictive models. The technology, observational strategies, and international cooperation are in place to enable ACE-Asia to make large gains in this regard.

II. Program Rationale and Scientific Questions

It is now widely accepted that aerosols cause the largest uncertainty in modeling the radiative forcing of climate (IPCC, 1995). The indirect (cloud) effect is generally thought to be the most uncertain, due to our inability to adequately describe the complex microphysics of cloud formation and the ways this is modified by aerosols (NRC, 1996). Although the direct effect of aerosols is considered to be more certain, even the sign of this effect is now in question due to the impact of absorbing aerosols (Sokolik and Toon, 1999). Sulfate aerosols have a cooling effect, but absorbing aerosols such as BC and dust (both major components of the Asian aerosol) may cause warming, depending on the albedo of the surface, their altitude, and even their latitude. In major parts of the globe their interactions with solar and infrared radiation are so similar in magnitude (but opposite in sign) that even the sign of the modeled direct forcing changes with very minor changes in assumptions about dust particle size (Woodward and Roberts, 1999). Unfortunately, there are virtually no observations with which to constrain such models. We hope to improve that situation.

Climate models often use a small subset of parameters to describe direct aerosol impacts (Charlson et al, 1992). The optical depth, τ , quantifies the total extinction by a column of aerosols, and includes the effects of aerosol concentrations. Modeling it thus requires predicting the concentration of various aerosol types over time and space (Langner and Rodhe, 1991). The fraction of the extinction that is due to scattering (as opposed to absorption) is described by the single-scatter albedo, ω_0 . A value of 1.0 means no absorption, while 0.8 (a value typical of some dust) means that 20% of the extinguished radiation was absorbed. Single-scatter albedos are intensive properties of various aerosol types, which do not depend on concentration. Values less than 1.0 imply heating of the absorbing aerosol layers. The third parameter is a phase-function, P that defines the angular dependence of scattering. This complex function depends on the refractive index of aerosols, their size distribution, and the wavelength of the incident light. It is often replaced by a simplified backscatter coefficient, g , to approximate the fraction of scattered sunlight that will be directed away from the Earth. Since most aerosols adsorb water at higher relative humidities, their size and scattering efficiency varies with RH. This is frequently expressed as a (strongly composition dependent) function, $f(RH)$, that describes the change of scattering with humidity for each type of particle.

The net effects of these parameters are sometimes expressed as mass scattering efficiencies for various components that can then be multiplied by concentrations to derive the extinction. Modeling the direct radiative effect thus requires modelers to accurately predict the variation of complex, time-varying, size-dependent chemical mixtures throughout the atmosphere. Needless to say, this is a Herculean task; in most parts of the atmosphere (especially above the surface) there are no data against which to test these models.

Chemical transport and radiative transfer models generally treat the tropospheric aerosol as several separate components; often mineral dust, black carbon, organic carbon, sulfate, industrial aerosols, and sea-salt (Haywood et al., 1999 and references therein). The various components have different mass scattering efficiencies, so the relative mass concentrations of the components and their size distributions will affect the aerosol phase function, the aerosol hemispheric backscattered fraction, the single scattering albedo, the wavelength dependence of scattering (Angstrom exponent), and the change in scattering properties due to particle hygroscopic growth. As all these variables are prescribed quantities in radiative transfer models; measurements of them in the ACE-Asia region will help to improve the parameterization of aerosol properties in this region.

The interaction between these species can affect their optical properties. Results from ACE 1 have shown that the atmospheric aerosol, even in remote regions, is rarely present as a pure chemical external mixture in any size range. In the ACE 1 study area, between 11 and 46% of the sulfate particles with diameters > 100 nm contained soot (Postfai et al., 1999). Organic species were detected in over 50% of the particles with diameters > 160 nm and were associated with sea salt (Middlebrook et al., 1998). Over 90% of the aerosol particles with diameters > 130 nm contained sea salt (Murphy et al., 1998). The degree of mixing of the various chemical species has a major effect on the optical properties of the aerosol. The presence of less soluble organic species will alter a particle's hygroscopic properties (Zhang et al, 1993). Similarly, mixing with soot will increase light absorption and decrease the solubility of sulfate particles.

Gas-phase and aerosol-phase chemistry are linked in ways we cannot now quantify. By its mere presence, the atmospheric aerosol exerts some influence on gas-phase tropospheric chemistry, both as a possible source of halogen oxidants (Sander & Crutzen, 1996) and as a sink for reactive trace gases (Sievering et al., 1992; Dentener et

al., 1996). Several current regional to global-scale modeling studies hypothesize what this effect might be, but no in situ measurements exist to constrain these hypotheses. Many aerosols grow by the condensation of gas phase species; what role does the gas-phase chemistry of the Asian-Pacific region play in influencing aerosol properties and evolution? With the presence of sea salt, mineral dust, and anthropogenic pollution aerosols, the western Pacific offers the opportunity to observe the influence of this Asian multicomponent aerosol on tropospheric chemistry.

Asian aerosols are also of interest because the region's emissions are changing due to rapid industrial development (van Aardenne et al., 1999) and increasing numbers of automobiles (Elliott et al., 1997). These changes in emissions will no doubt cause changes in aerosol populations. One of the most reliable ways to test our models of aerosol formation and removal is to examine their sensitivity to an emissions change. We should not miss this chance to perform an experiment of opportunity by studying the Asian outflow carefully now and again in several years. We rarely have a chance to observe geophysical systems before and after a change, yet it is an extremely powerful way to test our understanding. It is worth noting that the emissions change may not be what many expect: China is enforcing new emissions standards on both new and old vehicles that may make earlier projections obsolete.

Finally, the Eastern Pacific atmosphere has been one of the cleanest on Earth due to the size of the ocean, the active wet removal of soluble species, and the relatively low upwind inputs of aerosols and aerosol precursor gases. However, based on analysis of data from the Pacific Exploratory Missions (PEM) and ACE 1 over the period 1991-1996, Thornton et al. (1999) concluded that anthropogenic sources in eastern Asia now dominate the sulfur chemistry in the lower troposphere of the western North Pacific eastward from the Asian continent for more than 1500 km and substantially further in the mid and upper troposphere. In addition to sulfur, the mineral dust transported from Asia can dominate the aerosol optical depth over the North Pacific (Husar et al., 1997). In situ observations on the west coast of North America confirm that transport of significant amounts of mineral dust and pollutants does occur across the expanse of the Pacific Ocean basin (Jaffe et al., 1999). Although perhaps half the dust is natural, the remainder of this aerosol is being added to a region that has historically had an extremely low optical depth. Thus, the relative impact of emissions is much greater than it would be in less pristine regions. It behooves us to know the extent of the Asian impact and the scales on which this material is removed from the atmosphere.

II.A. Mineral Dust

Several issues make it hard to reliably predict the impact of mineral aerosol (Sokolik and Toon, 1996). Absorption and scattering depend strongly on the altitude distribution of the dust; it may serve as an alkaline reaction surface under certain conditions, and its size can change with time due to sedimentation. The difficulty of sampling large particles without artifact may have compromised many earlier observations.

Optical properties: Although it is known that the wavelength-dependent refractive index of dust varies strongly from one source to another (Saharan dust is reddish-brown while Asian dust is more yellow), the only data available to modelers now is from a few Saharan samples run long ago. Mineral dust originating in Asia should be less absorbing at UV and visible wavelengths relative to the Saharan dust because of lower concentrations of iron oxides (Sokolik and Toon, 1999a,b). Additionally, dust particles internally mixed with soot, sulfates, nitrates or aqueous solutions will have drastically different properties from those at the dust source (Levin et al., 1996). Aggregation of dust with soot particles would cause a decrease in the single scattering albedo, enhancing the heating effect (Sokolik, 1999b). In contrast, the single scattering albedo of dust-water and dust-sulfate particles increases as the fraction of dust decreases, resulting in less heating. There are few if any data on the mixing state of Asian multicomponent aerosols (MCA) with which to select candidate mixtures for use in models.

In our present state of knowledge we are unable to reliably predict where scattering or absorption dominates and by how much. Of particular concern are model studies that show a huge impact of the altitude of absorbing aerosols on TOA radiances (Quijano et al., 1999). An absorbing layer below clouds may have little effect (the albedo will be controlled by the bright clouds above), whereas a dust layer above clouds can sharply reduce the radiation scattered to space. This impacts the radiation budget, the ability to infer things like ocean color and optical depth from satellite sensors (Gordon, 1997), and dynamics: absorption heats dusty atmospheric layers at the expense of the surface and the ocean. The potential for significant feedbacks demands that we learn to measure and model this MCA reliably. Lidar observations (Murayama et al., 1998; <http://info.nies.go.jp:8094/kosapub/>) have shown that dust is frequently highly layered, sometimes reaching far greater concentrations aloft than at the surface. What causes this layering and how do the physical and chemical properties of mineral aerosol vary within and between these layers? In situ observations are lacking.

What is $f(RH)$ for the various aerosol types found in Asia? Hanel (1976) showed that Saharan mineral dust, once coated with sulfates and nitrates, can take up substantial amounts of water when RH is above 50%. This

situation clearly prevails downwind from desert and loess plateau regions of Asia; e.g., Parungo et al. (1997) showed that 50% to 80% of the >2000 nm diameter dust aerosols at Qindao were coated with sulfate. Thus, we expect $f(\text{RH})$ measurements to show that substantial water amounts are associated with coated mineral dust, especially once the modified mineral dust transports over the western Pacific Ocean. Changing composition with dust age will cause $f(\text{RH})$ to change with time, making it very hard to predict. Measurements are lacking with which to constrain $f(\text{RH})$.

One of the major conclusions from TARFOX and ACE-2 is that the widely used Mie theory for scattering of light by particles may often be misleading (Hoyningen-Huene, 1999). Even seasalt particles may sometimes not be perfectly spherical, so that the amount of scattering may be poorly predicted by Mie theory. The insoluble crystalline core of the Asian MCA may pose serious challenges in modeling light scattering.

Chemical issues: Mineral aerosols can provide surface areas for atmospheric reactions (Anderson et al., 1996). Long-range transport of mineral dust from the Asian continent may have a significant effect on chemical reactions associated with the conversion to sulfate (Figure 1) and nitrate. Prospero and Savoie (1989) analyzed data on islands in the central Pacific Ocean and found that aerosol sulfate and nitrate exhibit a strong seasonal cycle with peaks in the spring, coincident with dust transport, even though sulfate and nitrate are not associated with the mineral dust at the source regions (Inoue and Yoshida, 1990). In analysis of aerosol composition at Cheju Island, Chen et al. (1997) found different species exhibit different seasonal cycles, with sea-salt components showing peak values in winter, while non-sea salt (nss) components nss-calcium, nitrate, nss-potassium, and to a lesser extent, sulfate and ammonium, having higher values in spring. Carmichael et al. (1996) showed that sulfate and nitrate at Cheju Island are associated with mineral dust; Clarke and Porter (1991) observed this to be true at MLO. Thus, a variety of ambient measurements suggest that primary aerosols associated with continental outflows, and especially mineral dust aerosols, serve as sites for gas-to-particle conversion, influencing the partitioning of volatile species between the gas and the condensed phases.

Whether substantial sulfate will be generated in the mineral dust/pre-existing sulfate/water mixture is a difficult question to answer. Residual alkalinity is likely to be present in the mineral aerosol since China dust is 200-250% enriched in Ca (as CaCO_3) over that of world average soil/dust (Wen et al., 1996). However, it is not known how much of this CaCO_3 will be available alkalinity in the dust/sulfate/water mixture. Previous work by Dentener et al. (1996) and Xiao et al. (1997) assumed it was all available, but it is quite possible that only a small fraction is.

That global three-dimensional tropospheric model study by Dentener et al. (1996) suggested that the reaction of SO_2 with calcium-rich mineral dust plays an important role in SO_2 conversion downwind of arid source regions, like Asia. Model calculations by Xiao et al. (1997) also suggest that chemical conversion of SO_2 to sulfate in the presence of mineral aerosol may be particularly important in early spring when gas-phase reactions are comparatively slow, and may contribute from 20 to 40% of total sulfate production at that time. Their calculations are consistent with observations in the Pacific region. As the plots in Fig. 2 show, carbonate is displaced as aerosols react with acidic species. The greater surface to volume ratio in fine particles causes them to be depleted more rapidly than coarse particles. Single-particle analysis on Asian dust found that 50% to 80% of the large particles ($D_p > 2 \mu\text{m}$) were coated with sulfate (Parungo et al., 1995). Dentener et al. (1996) estimated that during the months of FMA, the region where over 40% of the total nitrate is found on mineral dusts was predicted to cover almost all of Asia and extend throughout the central and northern regions of the Pacific Ocean. They also suggested that interactions of N_2O_5 , O_3 and HO_2 radicals with dust affect the photochemical oxidant cycle, with ozone concentrations decreasing by up to 10% in and near dust source areas. The impact of these processes depends on the assumption that the alkalinity from mineral aerosols exceeds the acidity from sulfuric acid, sulfates and other acidic compounds associated with the MCA so that the water's pH remains well above 6. This assumption has not been verified by aerosol measurements in ACE-Asia region aerosols.

Size evolution and lifetime issues: The lifetime of mineral aerosol is a particularly interesting issue, since it will limit the lifetime of any adsorbed constituents. Near its source, the volume size distribution of dust may peak at 4-9 μm diameter (Parungo et al., 1997). Such large particles should have settling velocities of about 2 cm/s, which means that particles in a layer at 3000 m altitude should reach the surface in just 40 hours. At 7 m/s (Merrill et al., 1997), this means the particles should not travel more than about 1000 km. Yet Betzer et al. (1988) found 200 μm Asian dust particles in the mid-Pacific Ocean, some 10,000 km from their source! How do particles remain suspended for such long-range transport? Sedimentation theory (Windom, 1969) suggests that layers should become size-segregated during transport, with larger particles sinking as smaller particles remain aloft. Does this in fact happen? Westphal et al. (1987) predicted that sedimentation should deplete elevated layers of large particles and create lower layers enriched in the largest sizes. There should thus be less altitude variation of size close to sources than farther out at sea. The lifetimes and spatial impacts of mineral aerosols are no doubt extended when they are mixed into the free troposphere. There are presently no data with which to test these hypotheses.

This deposition has a biological consequence: dissolved iron limits biological productivity in much of the Central Pacific Ocean (Duce and Tindale, 1991). Since deposition of Asian dust is virtually the only source, it plays a crucial role in controlling such diverse factors as fisheries productivity and the burial of atmospheric CO₂. Dust removal rates and chemical processing during transport (solubilizing the mineral iron by adsorbed acids) control the biological impact on various regions of the ocean. The lack of observations of changes in size and chemistry with distance from dust sources inhibit the development of reliable iron biogeochemical models.

It is clear that mineral dust plays a very significant role in controlling the radiative, chemical, and biological environments over and in the Pacific, but very few data are available to test hypotheses concerning those effects. This seriously limits our ability to create and evaluate quantitative models of climate and associated impacts.

II.B. Carbonaceous Aerosols

Among the major issues unresolved by earlier experiments is the role of carbonaceous aerosols. It is something of an embarrassment that in most atmospheric reservoirs we cannot even specify the *fraction* of the aerosol that is organic (Hering et al., 1990), let alone provide critical information about its mixing state, functional groups, water solubility, impact on surface tension (Saxena et al., 1995), or the molecular weight of the compounds involved. Since in some continental outflow, organic species may comprise half or more of the mass (Hegg et al., 1997), our ignorance about organic speciation prevents us from making reasonable estimates of the growth of aerosols with changes in humidity, $f(RH)$. This is a critical factor in models of both direct and indirect climate forcing.

Organic particulate matter is both emitted in particulate form (i.e. primary) and formed in the atmosphere from products of gas-phase photochemical reactions (i.e. secondary; Hildemann, 1993; Turpin and Huntziker, 1995). Table 1 lists classes of organic compounds that have been measured in atmospheric aerosols or are predicted to be present based on photochemical and thermodynamic arguments (Saxena and Hildemann, 1996). Both biogenic and anthropogenic sources contribute to primary and secondary organic particulate matter (Grosjean, 1992; Hildemann et al., 1996; Mazurek et al., 1997; Schauer et al., 1996; Rogge et al., 1993). The heavy reliance on biomass as fuel in Asia contributes to the relatively high levels of both soot and organic aerosols. Organics of biological origin, such as pollen, spores, and microbial products (e.g. endotoxins or lipopolysaccharides) are also found in the atmosphere (McLean et al., 1991; Young et al., 1998). Many organic compounds are found in both gas and particle phases (e.g. several polycyclic aromatic hydrocarbons, carboxylic acids, organic peroxides, and others). Their phase partition is a function of atmospheric properties, such as temperature and relative humidity, sorptive properties of the aerosols, and properties of the organic compounds themselves, such as vapor pressure and solubility (Pankow, 1994). The phase partitioning of monocarboxylic acids is also strongly pH dependent (Keene et al., 1995). Since precipitation pH over much of China is above 6, "excess alkalinity" may cause these acids to reside primarily in the condensed phase. The presence of multiphase (e.g. semivolatile) organic compounds complicates the collection of organic particulate matter because the equilibrium between the gas and particle phases is often disturbed during sample collection (Turpin et al., 1994).

While in most field studies speciation of inorganic aerosol components such as sulfate and sea salt has been done (e.g. Quinn et al., 1996; Huebert et al., 1998), few measurements exist on organic aerosol speciation and even fewer on the altitude variability of organic aerosols (Kawamura et al., 1996; Novakov et al., 1997; Middlebrook et al., 1998). In one major field program (TARFOX) it was found that levels of organic aerosol aloft exceeded those at the surface (Novakov et al., 1997). Although this conclusion emerged as a principal result of TARFOX, the data on which this finding was based were lacking in many respects. Organic aerosol percentages of total aerosols measured at the surface in ACE-2 agreed more or less with those observed at the surface in TARFOX, although comparable measurements aloft were not taken.

Black carbon is the other form of carbonaceous aerosol that has a major radiative impact (Haywood et al., 1995). It absorbs radiation more strongly than dust, thereby causing similar radiative and dynamical impacts as dust. The radiative impact of BC is increased when it is associated with sulfate or other hygroscopic species (Posfai et al., 1999). The distribution and mixing state of BC in Asian outflow is not well known.

What controls the vertical distribution of organic aerosol and BC in the atmosphere? Several factors might lead to enhanced levels of organic aerosol aloft: (1) clouds and subsequent wet deposition remove sulfate more efficiently than organics, so cloud-processed air parcels might be expected to have organic aerosol levels preferentially enhanced; (2) lower temperatures aloft can lead to enhanced condensation of organics; (3) stronger UV aloft can lead to accelerated photochemistry, which might lead to accelerated production of secondary/condensable organic aerosol aloft; (4) hydrocarbons that are precursors to secondary organic aerosols can be advected aloft where they are oxidized to produce semi-volatile products; and (5) organic aerosols accumulate in layers aloft as a result of long distance transport from source regions. A prerequisite to determining the importance

of these and other possible factors is reliable aerosol measurements as a function of altitude that can be analyzed in terms of EC/OC ratio and key chemical species.

The relative contribution of primary and secondary sources to the total organic content of atmospheric aerosols in general, and particularly in the ACE-Asia region, remains largely unknown. Since this material plays such a major role in the uptake of water by particles, the usefulness of indirect and direct forcing models will be limited by their ability to treat organic matter realistically.

II.C. Sulfates and other anthropogenic/industrial ions

It is essential that models correctly predict the fate of SO_2 , since Thornton et al. (1999) have shown that high levels of SO_2 are present in the outflow from Asia. Its four most likely fates (dry deposition, homogeneous oxidation, oxidation in cloud, and oxidation in seasalt aerosol water or on dust) all have dramatically different impacts on the size distribution, amount, and lifetime of the resulting sulfate aerosol. Which of these processes dominates over land and which after the air has moved over the ocean? While there is general agreement that direct loss to surfaces (dry deposition) and conversion to sulfuric acid are the principal sinks for SO_2 , there is no consensus on the relative importance of these loss mechanisms on global or regional scales. The impact on the size distribution of homogeneous oxidation by OH (Cox & Shepard, 1980), in-cloud oxidation by peroxide (Penkett et al., 1979), and seasalt oxidation by ozone (Sievering et al., 1992) are all very different. Only the first can nucleate new particles and create accumulation mode sulfate. Do the relatively high levels of ammonia in Asian air (Zhao and Arpu, 1994) change the relative reaction rates by raising the pH? Observations are lacking to constrain models of these processes over the northwestern Pacific.

Gas-phase tropospheric oxidation is likely dominated by initial reaction with OH, but additional oxidants such as O_3 , NO_3 , and halogen species could also be important under some conditions. In the region downwind of major Asian industrial emissions, we would expect that high mixing ratios of NO_x and NMHCs, under the influence of light, will generate high O_3 mixing ratios (Jaffe et al., 1996; Akimoto et al., 1996). This elevated O_3 , combined with high relative humidities and strong solar actinic fluxes, make an ideal environment to generate significant mixing ratios of OH. At night, high O_3 and NO_2 mixing ratios could generate NO_3 radical, providing a further route for gas-phase oxidation. In addition, nighttime production of N_2O_5 and its subsequent reaction on wet aerosol surfaces may be a significant sink for reactive N, a source for particulate NO_3^- and acidity, and a source for ClNO_2 (a precursor for atomic Cl) (Keene et al., 1998).

The important role of clouds as efficient chemical reactors, especially for SO_2 , is well established. Increasingly sophisticated modeling studies of MBL stratiform cloud droplet chemistry suggest the S_{IV} + ozone pathway dominates H_2SO_4 production in this abundant and climatically significant cloud type (Ayers and Larson, 1990; Hegg et al., 1992). However, none of these modeling efforts appears to have considered the possible acidification of cloud droplets by HCl, HNO_3 , H_2SO_4 , HCOOH and/or CH_3COOH , all of which are probably present at high concentrations in the MBL downwind of Asia.

Mineral aerosol, which is generally alkaline, can also provide a reaction surface (Dentener et al., 1996). While in the free troposphere dust particles will generally be devoid of adsorbed water; in the MBL they will acquire a hygroscopic coating. Such water-coated mineral aerosols are known to provide reactive surfaces for both sulfate and nitrate production. Cations such as calcium play an important role in partitioning the semi-volatile components and thus in determining the size distribution of these anions. Carbonaceous aerosol may also provide sites for heterogeneous reactions. Some studies of sulfur and nitrogen oxides with carbonaceous particles show sulfate production to be more than 10 times greater than nitrate production (Mamane and Gottlieb, 1989). Yet a consensus of the global significance of this process is currently lacking.

Many uncertainties prevent us from reliably modeling the chemistry of sulfur (and nitrogen) oxides in Asian outflow. Since the size distribution of sulfate aerosols is so important for both direct and indirect climate forcing models, it is imperative that we learn which pathways SO_2 follows in various regions and altitudes.

II.D. Sea salt Aerosols

The extent of the impact of continental material is controlled by deposition processes. It is possible that much of the SO_2 that reaches the MBL is oxidized on seasalt and removed fairly rapidly (Sievering et al., 1999). The source and removal rates for seasalt aerosol have been parameterized by several groups (Gong et al. 1997; Erickson et al., 1999; O'Dowd et al., 1997). Yet even the MBL burden of seasalt is uncertain, due in part to ignorance of its vertical distribution: airborne measurements of Na^+ have been unreliable, since large particles are seriously under-sampled by aircraft sampling inlets (Huebert et al, 1990). The result is that the vertical profile of seasalt measured with an external device by Woodcock (1953) almost half a century ago is still one of the best in print.

The mass median diameter and sea salt mode mass/volume are strong functions of relative humidity

(McInnes et al., 1996). Three-fourths or more of the sea salt mode volume is water at relative humidities of greater than 80%, which are commonly observed in the MBL. This provides a significant water volume within the MBL for uptake and reaction of pollution derived gases such as SO_2 and HNO_3 (Sievering et al., 1992; Sander and Crutzen, 1996). Fresh seasalt aerosol undergoes rapid chemical transformations involving the scavenging of gases (e.g., SO_2), aqueous-phase reactions (e.g., oxidation of S_{IV} to S_{VI}), and the volatilization of products (e.g. various halogen species) (Vogt et al., 1996). Although these aerosols are initially alkaline, limited available evidence suggests that the seasalt alkalinity is rapidly titrated (seconds to minutes) by common acids in the MBL (e.g. HNO_3 , HCl , H_2SO_4 , HCOOH and/or CH_3COOH). Most ambient seasalt aerosols are probably acidic, with pH's ranging between 2 and 3 in polluted regions, and 4-5 in more remote regions. If so, the implications for SO_2 oxidation are significant since the mechanism generally believed to be most important-oxidation by dissolved ozone-is negligibly slow at pH below about 5 (Chameides and Stelson, 1992). Several recent modeling studies have suggested alternative mechanisms for S_{IV} oxidation by hypohalous acids in deliquesced seasalt (Keene et al., 1998). The presence of non-seasalt sulfur on seasalt aerosols probably does not affect the direct radiative forcing of these aerosols, but it does represent a sink for sulfur which might otherwise form new particles. Indeed, the nature and rates of many multiphase transformations are strongly pH dependent such that this parameter will be critical to monitor. There is no clear consensus on the relative importance of seasalt oxidation and cloud processing (Yvon and Saltzman, 1996) of SO_2 , although each will produce NSS (non-sea salt sulfate) of a very different size and lifetime.

It is now clear that the suite of important oxidants in the marine atmosphere includes halogen radicals. As an ozone-rich continental plume moves over the ocean (similar to the scenario modeled by Sander and Crutzen, 1996), chemical transformations involving sea-salt aerosol produce halogen-atom precursors such as Br_2 and Cl_2 thereby substantially altering oxidative processes based on conventional HOx/NOx chemistry. Different reaction pathways for sulfur, nitrogen, and carbon species in this modified oxidative environment impact atmospheric lifetimes of these species and, consequently, the rates at which condensable products are generated. In addition, halogen cycling strongly controls the pH of marine aerosol and thus important pH-dependent chemical transformations such as SO_2 oxidation in deliquesced sea-salt aerosol and the resulting particle size distributions of reactant species. Process studies (hopefully in concert with TRACE-P) are essential to determine how to most realistically model the oxidation state in the Asian plume.

Blake and Rowland's group found during ASTEX/MAGE that some NMHCs were lost faster than OH alone could explain (Wingenter, 1996). It is now possible to measure BrO (using DOAS) and Br_2 (with CIMS). If the model of Sander and Crutzen (1996) is correct, Br_2 should accumulate at night and disappear in the daytime, while BrO gets replenished only in the daytime and runs out at night. This chemistry may reduce ozone concentrations by 6 ppb or more, and it limits DMS concentrations to a small fraction of their halogen-less values.

Although sea salt is an important component in MBL chemistry, we lack observations with which to constrain models of its processes. Fundamental information about its variation with altitude and its evolution during chemical processing are needed.

II.E. Aerosol Evolution

CTMs and GCMs use numerical representations of numerous chemical and physical processes to predict the changes in aerosol concentrations and properties with time. Consider the change in photochemical processes over a daily cycle. How do OH (Eisele et al., 1997), NO_3 (Chameides, 1986), and halogen oxidants (Sander and Crutzen, 1996) change with time of day? All should have large diurnal cycles and each may play a role in oxidizing precursor gases to condensable species. The plume from Asia is likely to be complex, though: How rapidly does the photochemistry respond to the presence of seasalt? Does the increased humidity in the MBL change the concentration of OH? Does the presence of a large particle surface area change the abundance of these radical species? How do the diurnal changes in process rates affect the usefulness of various time-averages?

Our interest, of course, stems from the fact that the aerosols themselves will evolve as a result of these processes. Cachier et al. (1986) found that the organic aerosol derived from anthropogenic gases increases with distance from Asia. Does the ratio of carbonaceous to NSS mass change with time in the Asian plume? It is conceivable that the sulfur is oxidized more rapidly, so that at some distance offshore continental sulfate concentrations are dropping as organic vapors are just being oxidized to a condensable form.

Physical processes will also cause an evolution of the aerosol. Presumably the MCA starts as relatively simple mineral particles, some of which may collect smaller aerosols by coagulation and sulfate (and organic species) by condensation of vapors. Such changes would cause both the single-scatter albedo and cloud-nucleating properties to change, so we need to be able to model them realistically. Does BL dust evolve more rapidly than FT dust? It may be that the different conditions in various layers require us to use very different approaches to describing the changes in each.

II.F. Layering and vertical transport

One of the most serious problems for modelers is the lack of aerosol data from layers above the surface. Layering is known to occur for many species in the free troposphere, in contrast to the gradual changes many models predict. Free-tropospheric temperatures, RHs, and aerosol lifetimes are very different from those at the surface, so reliable measurements of aerosol properties are essential to test models of aerosol processes and effects at various altitudes.

The vertical movement and exchange of airmasses play a huge role in the control of aerosol populations and properties. In the simplest sense, updrafts and downdrafts change the relative humidity that causes particles to gain or lose water. Clarke et al. (1998) and Raes (1995) argue that in the MBL particle number is controlled by subsidence from the FT. Russell et al. (1998) showed that the size distribution in marine areas is strongly impacted by mixing between layers. In an ASTEX/MAGE (Zhuang and Huebert, 1996) Lagrangian experiment, vertical mixing was the largest term in the ammonia budget. It is critical that we demonstrate an ability to accurately model the exchange of air, aerosols, and precursor gases between atmospheric layers.

Much of this transport is discontinuous: the lofting of dust from Mongolia and western China and its transport to the Pacific Ocean is not a "mean" process. It typically occurs in highly discrete events behind the cold fronts of developing midlatitude cyclones where surface wind speeds exceed the threshold needed to loft dust (Carmichael et al., 1997). These storms are frequent in the spring and are associated with an area of intense cyclogenesis known as the "Pacific storm track". Developing baroclinic waves vigorously mix tropospheric air, lofting dust, aerosols and trace gases into the free troposphere while simultaneously injecting stratospheric air around the tropopause folds (Rood et al., 1997). The jet stream in the storm track is very strong so anything lofted from the surface can be rapidly transported far down wind of Asia. Dynamic mixing at the storm track is probably a key factor for teleconnecting local Asian aerosol chemistry and dynamics to hemispheric scales. Rn-222 can be used as a proxy for continental emissions (Whittlestone et al., 1992) in studying such sources.

An initial investigation into the role of surface-stratospheric exchange around the Pacific storm track was performed with the 3D Geophysical Fluid Dynamics Laboratory Global Chemistry Transport Model (GCTM) for gas phase oxidant chemistry. The prominent role of transient eddies in transport out of East Asia results in downwind time series of CO mixing ratios (a proxy for continental emissions) that are quite noisy. For example, 1500 km downwind of Japan at the surface, the CO time series (Figure 3a) is influenced by significant and discrete transport events from Asia at a frequency of 4-5 per month. At 500 mbar, the CO time series (Figure 3b) is much noisier because of high frequency stratosphere-troposphere exchange in the heart of the storm track jet stream. The lofting around the Pacific storm track is what makes Asian aerosol and oxidant chemistry a hemispheric concern. A scatterplot of total CO vs. the Asian contribution to CO at 500 mbar in a GCTM grid box over the western US (approximately Montana) shows that nearly all high CO events are attributed to Asian CO. After lofting into free troposphere, the Asian CO (and presumably dust, aerosol, and other trace species) often traverse the Pacific in coherent parcels that elevate ambient levels over the US for 1-5 days. More detail can be found at http://www.cgrer.uiowa.edu/people/mphadnis/ace_asia.html.

There is also a practical reason for constraining mixing rates: Lagrangian observations (Huebert & Lenschow, 1999) are sometimes used to test our ability to model oxidation, condensation, and other process rates realistically. Since those processes are occurring in an environment in which mixing is also taking place, our ability to tease out the process rates is limited by our skill at quantifying the impact of vertical mixing on concentrations and aerosol properties (Zhuang and Huebert, 1996; Russell et al., 1998). Vertical exchange plays a major role both in controlling aerosols themselves and in our ability to understand them.

An interesting feedback may affect the rates of vertical exchange. Because the Asian MCA contains absorbing species (BC and mineral aerosol), the absorption of long- and short-wave radiation may cause plumes to become self-lofting. This behavior, which was noted in the dark Kuwait oil-fire plumes (Herring and Hobbs, 1994), may modify the dynamics of dust layers and increase the range of their transport. Understanding exchange rates between layers is every bit as important as knowing chemical rate constants for modeling the evolution of aerosols with distance from sources.

III. Scientific Objectives

The goals of ACE-Asia are to determine and understand the properties and controlling factors of the aerosol in the anthropogenically modified atmosphere of Eastern Asia and the Northwest Pacific and to assess their relevance for radiative forcing of climate. To achieve these goals, ACE-Asia scientists will pursue three specific objectives:

- Objective 1. Determine the physical, chemical, and optical properties of the major aerosol types in the Asian Pacific region, determine its state of mixing, and investigate the relationships among these properties.
- Objective 2. Quantify the physical and chemical processes controlling the evolution of aerosols in the Asian Pacific region and in particular their physical, chemical, and optical properties.
- Objective 3. Evaluate numerical models which will extrapolate aerosol properties and processes from local to regional and hemispheric scales, and assess the regional direct and indirect radiative forcing by aerosols in the Asian Pacific region.

In order to meet these objectives, we propose to collect data that can be used to initialize and test aerosol models for the region and measure process rates so these models can include more realistic depictions of the controlling processes. Numerical models are the vehicle through which all our knowledge of aerosol sources, processing, removal, and effects will be codified. Our observations are being designed with the help of modelers to ensure that they are effective for testing and improving process, regional, and hemispheric aerosol models. The Survey and Evolution Component (AA-SEC) will address a set of specific questions within these objectives which are described below.

Goals and Objectives of the Survey and Evolution Component

The questions AA-SEC proposes to address can be divided into three groups. The first is related to characterizing the concentrations of aerosols and their physical, chemical, and optical properties on several spatial and temporal scales that cannot effectively be accomplished by the routine surface network operations. These data will be used to initialize and test models of aerosols and their impacts. The second set of questions is directed at the processes controlling aerosol concentrations and properties, such as reaction rates and vertical mixing. These observations will be used to make more realistic parameterizations of processes in aerosol models. The last set of questions concern subtle feedbacks between aerosols and other atmospheric chemical processes, such as the creation of new oxidants from sea salt.

Aerosol Survey

What are the vertical and regional distributions of aerosol concentrations and properties under various meteorological conditions?

- A. What are the relative size-dependent contributions of sulfate, nitrate, carbonaceous material, sea salt, primary anthropogenic particles, and dust to the optical properties (absorption, total and angular scattering) of aerosols?
- B. How do the interactions between these species (their state of mixing) affect their optical properties?
- C. How are the distributions affected by specific meteorological events?

Aerosol Evolution

How do aerosol properties evolve as they move offshore?

- D. How do aerosol properties and process rates evolve with time (distance) over the ocean?
- E. What are the lifetimes or removal rates of particles?
- F. What is the rate of exchange of particles and precursor gases between the free troposphere, buffer layer, and boundary layer?

Aerosol Interactions with Gas-Phase Atmospheric Chemistry

What role does the atmospheric chemistry of the Asian-Pacific region play in influencing aerosol properties and evolution, and likewise, how does the Asian aerosol affect tropospheric chemistry in the region?

- G. How does mineral dust interact with atmospheric gas-phase chemistry?
- H. How does sea salt interact with gas-phase chemistry?
- I. How does the anthropogenic/urban aerosol interact with gas-phase chemistry?

These three sets of questions will guide our experimental design. In each case we will use a variety of models to interpret the data, thereby improving the models. For each question we have identified a set of tasks that will be used to reach answers.

IV. Experimental Design and Observational Requirements

IV.A. Tasks

Aerosol Survey Tasks

The Intensive Aerosol Survey is an extension of the network studies and will:

- Extend the network study into the vertical and out to sea, to enable measurements of aerosol and aerosol precursor distributions on many spatial and temporal scales. (e.g. tens of m to km in the vertical; km to hundreds of km from the coast; hours to weeks)
- Provide additional measurements that can not be performed on a routine basis to more fully characterize the aerosol (e.g. more highly size-resolved chemical composition, number size distribution, hygroscopic properties, extent of internal/external mixture, organic aerosol properties)

Aerosol Evolution and Interactions Tasks

By measuring the change in dependent variables along with a suitable set of independent chemical and physical variables, it is possible to quantify the processes responsible for the change. Comparisons of these observed relationships with model parameterizations will be used to make models more realistic. The aerosol evolution experiments will look for changes in aerosols, their precursors, and their properties with time by repeatedly sampling airmasses using three approaches:

- Surface sites along a wind vector or airmass trajectory,
- Arranging ships at different distances downwind of sources, and
- Conducting airborne Lagrangian experiments with multiple aircraft, ships, smart balloons, and chemical tracers.

IV.B. Measurements and Experimental Strategies

We will use two observational strategies to gather data for testing and improving models: 1) local closure experiments performed in a wide variety of places, and 2) Lagrangian experiments using marker balloons and tracers. Ideally, each intensive ground site (not including the many regular network sites), ship, and aircraft will have a suite of instruments that allow it to perform some closure experiments (Table 2).

Surface sites are needed to provide the physical space and longer sampling times for more detailed measurements (e.g. tandem DMAs to measure hygroscopic growth and $f(RH)$, multistage chemical impactors for more size resolution, lengthy sampling for gravimetric analysis, sampling for organic speciation). Our intention is that each surface site will have instruments for measuring precursor gases, chemical and physical aerosol size distributions, a suite of optical properties, and aerosol optical depth (Table 2). We propose that 4-6 of the more highly-instrumented network sites be run in an intensive mode during AA-SEC. Some more intensive efforts, such as those to characterize the mixing state of aerosols and speciate organic aerosols, will be done at only a few sites.

In both ACE-1 and ACE-2, homogeneity was sought. In ACE-Asia, however, we will be looking for differences. We want to measure process rates, concentrations, and properties of aerosols both within and adjacent to plumes. The idea is to keep everything constant but the aerosol and precursor concentrations. Ships can be located across gradients (Figs. 4 & 5) to gather simultaneous impactor samples. Aircraft can fly across the gradient or do upwind/downwind legs parallel to the interface. Some aerosols, like dust, come out in layers. How do the properties of aerosols change in, above, and below a layer? Looking at contrasts like these will help us to identify the controlling processes that link precursors and meteorological conditions to aerosols and their effects. We will be helped in finding these contrasting regions by real-time, operational 3-D modeling of aerosols by both D. Westphal (NRL) and C. Benkovitz (Brookhaven).

International collaboration: For US-NSF purposes, we describe below a minimum collection of US platforms (1 ship, 2 aircraft and 1-2 ground stations) that would constitute a core to address the issues above. However, the survey and evolution experiments would both be strengthened considerably with the addition of platforms from other nations: we could make measurements at more altitudes and locations simultaneously and provide more complete coverage of airmasses. With aircraft, for instance, there is always a question whether measured concentrations at several altitudes are different just because they were not sampled simultaneously.

As of this writing, there is a fairly strong probability that several other nations will contribute platforms. Possible aircraft include one from Australia (Siems), two from Japan (Kondo and Nakajima), one from Korea (Kim), and a leased Chinese aircraft (Wang). One ship each is likely from Japan (Uematsu) and Korea (Hong).

Measurement Needs:

The intensive measurement platforms must have the capability to simultaneously measure the aerosol

chemical, physical, and optical properties. The following instrumentation is the minimum requirement for state-of-the-art characterization of aerosol properties. The measurements listed in A and B should be included on the surface and airborne platforms:

- A. In-situ measurements:
 - 1. Chemical:
 - a) Total and size resolved chemical sampling for inorganic ions, mineral dust, and total and organic carbon. The number of stages will depend on the time resolution required. A minimum for all platforms is one size cut that gives a sub and supermicron fraction with a well-characterized upper size cut.
 - b) Single particle analysis by mass spectrometry and electron microscopy
 - 2. Physical:
 - a) Number size distribution from 5 to 10,000 nm diameter. The upper size cut will depend on the platform and inlet but must be well characterized.
 - b) Size resolved mass (gravimetric) size distribution (surface platforms only)
 - c) Total particle number
 - 3. Optical properties:
 - a) Total and sub-micron light scattering and backscattering by aerosols at 3 wavelengths
 - b) Total and sub-micron light absorption by aerosols
- B. Column measurements:
 - 1. Aerosol optical depth
 - 2. LIDAR measurements of aerosol vertical distribution at a network of sites
- C. Selected platforms should also include additional measurements of aerosol properties such as:
 - 1. Hygroscopic growth (tandem DMPS)
 - 2. Scattering and backscattering as a function of RH ($f(RH)$)
 - 3. Aerosol phase function measurements (from 3-177 degrees) at two or more wavelengths
 - 4. 180° backscatter (for lidar interpretation)
 - 5. Higher time resolution chemical analysis (total sulfur, carbon, nitrogen at 1-2 hour time resolution)
 - 6. Organic speciation and water solubility
 - 7. Mineral composition to distinguish reacted from unreacted mineral aerosol as well as identify source regions
 - 8. Aerosol precursor gas concentrations (SO_2 , DMS, NH_3 , HNO_3 , NMHC) to compare with aerosol distributions in chemical transport models

While it is desirable to have these additional measurements at as many sites as possible, from a practical standpoint regional measurements at a limited number of sites should be sufficient to characterize these properties.

There are several measurements that may be considered high risk (because they have not yet been described in peer-reviewed publications) that should be made a high priority because they could add so much to the experiment. This list includes a polar nephelometer (Porter et al., 1998) for measuring the aerosol phase function, an incredibly important property of aerosols that directly impacts climate models as well as our ability to measure aerosols by remote sensing and OPC techniques. Fast (>10 Hz) measurements of SO_2 and DMS also fall into this category: an APIMS instrument will be flight tested in the fall of 1999 and hopefully again in 2000. The ability to measure the flux of SO_2 by eddy correlation would add a very strong constraint to our Lagrangian sulfur budgets. Eddy correlation measurements of DMS flux are a high priority here less for their chemical interest than for their ability to tightly constrain exchange rates and the dynamics of the MBL (Huebert & Lenschow, 1999). Finally, porous diffuser inlets have only been lab tested as of this writing, but will be flight tested in 2000. They offer the potential to vastly improve our ability to sample supermicron particles from aircraft. In each case the potential payoff is so large that we strongly recommend putting these methods into use if at all possible.

Based on the priorities listed in Table 2 and considerations of space, power, and equipment availability, we have compiled recommended instrument packages for the C-130, Twin Otter, and NOAA/UNOLS ship in Tables 3, 4, and 5. Table 6 lists the needed numbers of flight hours and ship days.

IV.C. Survey Research Strategy

The sampling strategy during the intensive survey will be to obtain an over-determined data set, using multiple techniques to define the aerosol chemical, physical, and optical properties, so that we may test for closure. To support process models, we will also measure precursor gases, photochemical species, and natural and anthropogenic tracers that can be used to establish air mass histories.

Closure studies are a key concept behind integrating models and measurements (Quinn et al., 1996). In such an experiment an over-determined set of observations is obtained and the measured value of a dependent variable (e.g. measured light scattering by aerosols), is compared with the value that is calculated from the measured values of the independent variables (e.g. measured chemical and number size distributions), using an appropriate model (e.g. Mie or an alternate scattering model). The outcome of a closure experiment provides a direct evaluation of the combined uncertainty of the model and measurements. If there is agreement between the measured and calculated values within the accepted level of uncertainty, the model may be a suitable representation of the observed system and appropriate for use as a component in other higher order models. Poor agreement indicates that there are problems in the model or measurements that must be corrected before proceeding further. Quantification of uncertainty is essential for a meaningful closure experiment. We will seek closure for:

1. Aerosol mass as a function of particle size derived from the number size distribution, chemical analysis of aerosol species and gravimetric analysis.
2. Measured hygroscopic response of the aerosol size to changes in relative humidity (RH) and the hygroscopic response calculated from the measured aerosol number and chemical mass size distributions, RH and published functional relationships between chemical composition and water uptake.
3. Measured aerosol scattering and calculated aerosol scattering derived from Mie theory applied to measured number and chemical mass size distributions.
4. Measured increase in aerosol scattering due to hygroscopic growth and the calculated increase based on measured number and chemical mass size distributions, RH, and published functional relationships between particle chemical composition and water uptake.

The survey sampling plan can be divided into 3 types of operations: a general grid survey, a sea-salt gradient experiment, and a dust layer study. The three types of operations are described below.

Grid Survey

The intensive survey of aerosol properties can best be accomplished with a multi-platform approach. At least one aircraft is needed to obtain vertical distributions of aerosol properties. This plane would fly sets of vertical legs (a “wall” profile, Figure 4) on some flights and horizontal gridded patterns on other flights (Figure 5). Several possible wall profiles are shown in Fig. 4, which could be flown sequentially or (preferably) by several aircraft simultaneously. Depending upon the meteorological conditions the ship could be positioned to sample different air masses and aerosol types. Multiple ships, positioned at various distances offshore, would allow simultaneous measurements of aerosol properties along a trajectory and thus the opportunity to assess the change in aerosol properties with distance from shore.

Sea Salt Gradient Study

Sea salt aerosol is important in radiative transfer (Haywood et al., 1999) and an important and reactive surface for atmospheric chemical reactions (Sievering et al., 1992). How does the concentration and size distribution of sea salt change with altitude? Are the larger particles more concentrated near the surface, due to the effect of sedimentation? Is it less heavily processed when it has just been emitted, but more acidic after aging with continental air? Is the bromine more depleted in the upper buffer layer (BuL) than in the boundary layer (BL)? The objective of this set of observations is to characterize the physical and chemical size distributions of the sea salt aerosols in the surface mixed layer.

The circular patterns and porpoising across the inversion by the C-130 (Figure 6) are intended to quantify the dilution of the BL air by BuL air, since that may have as large an effect on the size distributions as settling and surface production. Ideally, the ship would be in the exact middle of the first circular leg, so that the chemical size distributions would be taken from the average air that was sampled by the aircraft. The Twin Otter would collect hour-long Moudi samples at several altitudes and then do a stair-step sounding that will allow nephelometers and size-resolving devices to stabilize and average for a few minutes at each altitude.

Alternatively, one aircraft could also do a few long samples, identifying an altitude of interest and staying in it (perhaps flying racetracks) for 2 hours or more, to gather a large sample for organic or trace metal analysis. As unexciting as it may sound for many instruments, size-segregated organic composition is so important that some concentrated flight hours must be devoted to it.

Sea salt aerosols are important reactors for marine boundary layer chemistry. By devoting two flights each of the C-130 and the Twin Otter and a couple of ship-days, we could increase our understanding of this chemistry tremendously.

Dust Layers Study

This study will be designed using input from the lidar network (Fig. 7, to obtain both spatial variability and vertical structure) and satellite observations (to obtain horizontal extent) to identify locations of maximum dust concentrations and areas where concentration gradients can most easily be studied.

After the ferry to this region (Figure 8), the first leg for the C-130 would be a high-altitude lidar leg of about an hour's duration to map the altitudes and extent of the dust plume. The hour could be linear flight (either across the plume or down its axis) or doubling back and forth to map the width at various distances from the coast. It would provide the information for planning the legs in each of several layers. After the lidar leg, a slow sounding would permit in situ measurements of light scattering and other quickly-measured properties, to help narrow down the choice of altitudes to study. There are several ways a layer could be studied, and three are illustrated here. The various times would allow for greater altitude definition by short legs or longer legs to support longer collections for organic analysis. The Twin Otter is shown sampling both in and between layers to collect Moudi samples.

Since the horizontal extent of layers may be difficult to gauge, the approach at 4200 m might be useful: porpoise through the layer to see if it rising, descending, or fading away along one direction, and then plan a very long sampling leg based on that information. During INDOEX and PEM-Tropics B, aircraft were able to remain in layers for extended periods of time based on modest porpoising to seek and remain within the center of a plume. A ship would be valuable in this scenario for characterizing both the time-series of this event (using both lidar and in situ measurements) and for collecting more detailed chemical size distributions than is possible from the aircraft. In view of the difficulty of defining such large features, extra aircraft would add tremendously to the science. That kind of parallel study would benefit from the careful measurements of optical properties and surface chemistry in each layer.

IV.D. Aerosol Evolution Research Strategy

To measure the evolution of aerosols and precursors one needs to observe the same air mass over time. In some cases, a few hours might be enough to see significant changes, as in the case of hydrocarbons measured by Wingenter et al. (1996) in ASTEX/MAGE. In other cases, it is desirable to observe a full diurnal cycle, including an entire day's photochemistry (starting before sunrise), to record the rise or fall of species as the sun light comes and goes. While it is not possible to observe a totally isolated air mass over time (mixing will occur at the boundaries), by constraining much of the dynamics, one can solve for process rates.

The difficulty, of course, is knowing that one is sampling the same air mass at different points in time. There are several ways this can be done. One is to position two ships upwind and downwind of one another, as in Figure 9. The arrows represent mean springtime winds east of Asia. To assure that the two platforms are sampling the same air mass in such a heterogeneous environment, a balloon and/or chemical tracer can be released from the upwind ship.

Ground stations also can be used in a similar fashion. To study sea salt impacts on the SO₂, for example, it may be possible to align Qindao, Cheju, and a downwind ship along a forecast air mass trajectory. When conditions seem appropriate, a balloon could be released from the upwind site to tag the air mass and verify that the 2 or 3 other sites are indeed situated along a streamline.

Airborne Lagrangian experiments: One of the most elegant approaches to Lagrangian observations is to tag an air mass with inert tracers and instrumented balloons, so that aircraft can repeatedly sample the full vertical extent of the moving column (Zhuang and Huebert, 1996; Bates et al., 1998; Suhre et al., 1998). Huebert and Lenschow (1999) discuss the development of this observational strategy. In such an experiment a considerable effort has to be applied towards measuring rates of exchange between layers (Russell et al., 1998). It is this exchange that limits our ability to ascribe observed concentration changes to chemical processes or surface gain and loss. The better one measures relevant dynamic factors such as entrainment rates, the more precisely one can quantify reaction rates.

This type of Lagrangian experiment depends on the aircraft flying legs (usually circles) at a variety of altitudes. Each "stack" (Figure 10) is oriented the same way relative to the balloons that are tagging the air mass. The usefulness of the Lagrangian observations is maximized when the study air mass is under continual observation, which requires a relay of aircraft. The example in Figure 11 illustrates one possible Lagrangian experiment, using the NCAR C-130 and the CIRPAS Twin Otter flying out of Fukuoka, Japan. The basis for this example is trajectory analyses run on analyzed winds from the first week of March, 1997. Details can be found on Karsten Suhre's web site, <http://www.aero.obs-mip.fr/ace/ace3/trajs/>

At the start of the experiment, when the air mass is closest to the airfield, the ferry to and from the air mass takes just 3 hours (assuming 100 m/s airspeed), leaving 6 hours on station from a 9 hour flight of the larger aircraft. At this point the two aircraft could alternate 5-6 hours on station and cover virtually 100% of the time. However, as

the airmass moves farther from the airfield, the ferry time consumes a greater fraction of each flight, leaving fewer hours to do stacked circles in the target airmass. Obviously, more aircraft and greater endurance are desirable to maximize the coverage of the airmass. The ability of Lagrangian observations to constrain models is reduced considerably by long periods when no aircraft or ship was in the airmass (Suhre et al., 1998).

During these Lagrangian observations we will focus on the size-dependent chemistry, as a way of identifying changes that can be ascribed to particular processes. (Seasalt aerosol water SO₂ oxidation is the most likely source of increased NSS in the supermicron mode, for instance.) This set of measurements will also support modeling of potential heterogeneous reactions, condensation rates, and other size-dependent phenomena. Although we are more familiar with the possible mechanisms for sulfur oxidation, the same strategy will be informative for identifying the mechanisms by which carbonaceous aerosols are formed and modified. The evolution of sulfate size distributions and gas phase concentrations with time can help us quantify the amount of SO₂ that follows each pathway.

Budgets and dynamics: In a Lagrangian air parcel, the budget of any species, *S*, can be described by a continuity equation (Lenschow et al., 1988; Kawa and Pearson, 1989):

$$d\langle S \rangle / dt = J_0(S) - J_h(S) + F(S) - D(S)$$

Entrainment fluxes into the MBL, *J_h(S)*, can be parameterized in terms of the entrainment velocity (*ω_e*, common to all species being mixed) and concentration differences across the capping inversion. Repeated measurements in the volume can define *d<S>/dt*, the time-rate of change for each species. If the chemical formation and loss rates, *F(S)* and *D(S)*, of this substance can be measured, the surface flux, *J₀(S)*, may be solved for as a residual. Alternatively, if the surface flux is measured, it is possible to solve for the net chemical formation/destruction rate. In either case, however, the largest uncertainty is likely to be related to the entrainment rate measurements. To date these fluxes have depended on flux measurements of ozone, temperature, and water vapor, gradient measurements of aerosol size distributions and DMS, and wind divergence measurements (Russell et al., 1998; Lenschow et al., 1999). None has been shown to be accurate in all circumstances.

An ideal molecule for quantifying vertical exchange in the MBL is DMS (Huebert & Lenschow, 1999). DMS comes only from the surface (so there are virtually always quantifiable differences between layers), it is negligibly soluble in clouds, and it has a photochemical lifetime of a few days so it decays measurably in layers not in contact with the surface. If we could directly measure the DMS flux by eddy correlation, we could directly calculate entrainment velocities between layers. Other species fluxes could be accurately calculated by analogy, using the DMS-derived *ω_e* and easily-measured concentration differences across inversions. The challenge, then, is to develop a fast (10 Hz) DMS system for directly measuring the DMS flux from aircraft. Two fast DMS techniques offer promise: fluorine-induced chemiluminescence and atmospheric pressure ionization mass spectrometry; but neither has been demonstrated in the field. Both should be given a high priority for developmental assistance, since they would markedly increase the accuracy of Lagrangian-derived process rates.

Free tropospheric evolution: No one has demonstrated the ability to conduct a Lagrangian experiment in a FT air parcel. However, in several experiments aircraft have followed a layer hundreds of km from its source (most recently in PEM-Tropics-B, A. Clarke, personal communication). While this is not a strictly Lagrangian approach (any one part of the airmass is sampled only once), if the source of the layer is well known (in PEM-Tropics it was outflow from a cloud system), greater distances from the source imply longer lifetimes and the evolution that causes. After suitable corrections for the different photochemical conditions that each age of air may have experienced, it is possible to draw some conclusions about process rates.

We propose to try following elevated dust layers over the Pacific. If a particularly strong one passes over Cheju Island during our deployment, we will launch one or two smart balloons (they now have the ability to receive commands from an aircraft so they can be moved into a particular layer) and try to intercept them on a later flight. The higher wind speeds of the FT, however, make this a high-risk venture; we may not have enough range to reach the balloons for a later flight. It is worth the expenditure of a balloon to try, though, both to test the Lagrangian strategy in the FT and to have an opportunity to see whether sedimentation modifies the dust size-distribution in a predictable way.

Removal strategies: Potential new observational strategies for ACE-Asia include satellite observations of the decay of backscatter by the continental plume, Lagrangian in situ observations of concentration changes, eddy correlation measurements of dry deposition (e.g. for SO₂), collections of precipitation, and measurements of seasalt aerosols (and related buffering capacity) at several distances from the coast by ships and at island sites.

Since the value of a Lagrangian data set increases as the coverage becomes more complete, using two or more aircraft with significant range would add tremendous value to the data set over what could be derived from a

solo aircraft.

Aerosol Evolution Measurement Needs: An airborne Lagrangian experiment is a complex undertaking. It requires:

1. The infrastructure to identify follow-able airmasses and an operations center to coordinate the many activities
2. Smart balloons and chemical tracers to tag the airmasses
3. At least one ship to launch balloons, make longer collections, and measure surface water concentrations in support of air-sea exchange aspects; two ships add tremendously to the science by providing the capability to collect more precise chemical size distribution data at both ends of the trajectory
4. At least one long-range, heavy-lift aircraft (double crewed) and preferably one or more additional aircraft to maintain complete coverage of the airmasses
5. Instrumentation to measure a comprehensive suite of aerosol and precursor concentrations and properties

Infrastructure: Not all airmasses are good candidates for Lagrangian experiments. To observe the evolution of an airmass and have a reasonable chance of measuring process rates, the air has to be moving at a speed and in a direction such that aircraft can reach it and have several hours on station to study it over a period of at least a day. Thus, high winds and trajectories that pass over islands are to be avoided. Excessive vertical mixing due to convective activity is also a problem, since it destroys the integrity of the airmass and introduces dynamic terms that cannot be well constrained. As it happens, in the spring this region had numerous trajectories that were followable. Figure 12 shows in red the trajectories in 1997 that could have been reached from Fukuoka with a C-130 on one day in the spring of 1997. Figure 13 demonstrates that on almost every day there were one or more locations from which a successful Lagrangian experiment could be started. Further information about the assumptions used in these trajectory calculations can be found at <http://www.aero.obs-mip.fr/ace/ace3/trajs/ver2.html>.

To identify these candidate trajectories in real time it is necessary to expend considerable effort to forecast the movements of airmasses. Ideally we would have several groups producing forecast trajectories, based on a variety of forecast wind fields. During ACE-1, for instance, both ECMWF and NMC forecasts were used to predict airmass trajectories for selection of study airmasses. The agreement or disagreement between the different models was useful information in making a selection. This process needs to include a team of weather forecasters, which must include experienced regional forecasters who are knowledgeable about the vagaries of weather patterns in the study area. It also must include real-time downlinks from satellites, to provide current observations of cloud fields and the regional weather. Ideally this processing would take place in an operations center that is physically located near the major aircraft base, so that the air crews and scientists could be intimately involved.

When trying to follow dust layers, observations from a network of lidars are essential to identify the horizontal and vertical extent of the dust. It is not a simple matter to coordinate a network of these complex instruments and make their data available in near-real time, but the payoff is huge both in terms of experiment planning (where to send ships and aircraft to get the best samples) and later (to ensure that in situ data is of maximum value for interpreting the lidar backscatter).

Since air crews and ships must be positioned to intercept the selected airmass, it is essential that a candidate trajectory be identified at least 24 hours in advance of the start of observations. Although the precision of forecasts improves as the forecast time decreases, this gives time to reposition the launch ship (which can only move about 500 km per day) and alert air crews to prepare for an intensive operation period. Fine-tuning of the trajectory and making a final go/no-go decision can take place right up until the first flights or balloons are launched. The speed of the launch ship limits the ability to make last-minute changes.

Once the experiment commences, its course is dictated largely by nature: with the aid of the smart balloons, chemical tracers, satellite observations, in-situ observations, reliable communications links between all participating platforms, and around-the-clock forecast support, a relay of aircraft and ships observe the target airmass for as long as it is coherent and reachable. This requires a high degree of coordination and communications support from the operations crew.

Balloons and chemical tracers: The principal limit on the quality of the Lagrangian data is the extent to which the same airmass was observed throughout the experiment. This requires two kinds of tracers: balloons and chemical tracers.

Getting aircraft and ships back into the same airmass requires an active identification of the airmass by balloons that can float with the airmass and radio their location (Businger et al., 1999). It is now clear that these need to be “smart” balloons, so that they can detect the altitude change caused (for instance) by drizzle accumulating on the balloon, and compensate for this change in buoyancy. This is accomplished by pumping air into or out of a bladder that is inside a fixed-external-volume helium-filled balloon. This enables the balloon to remain in a boundary layer airmass and avoid either entering the ocean or drifting into the free troposphere. Smart balloons can

now also receive a signal from an aircraft, telling them to move to a different altitude, if wind shear, layering, or other considerations make it advisable. Low-power telemetry is used to broadcast the balloons' GPS-derived location and a variety of housekeeping and meteorological variables every few minutes.

The ideal experiment would involve releasing 5 or 6 balloons, so that dispersion could be calculated directly from the trajectories of the balloons. The technology for making and deploying these balloons has been improving with each subsequent Lagrangian experiment, but in practice we have worked with between 1 and 3 successful balloons. Protocols are established before the initiation concerning how aircraft will orient their sampling legs relative to the balloons.

Of course, wind shear in the boundary layer tends to smear out an airmass. In ACE-1 Lagrangian B the balloons spent much of one night near the surface, raising questions about whether their trajectory was slowed relative to that of the rest of the MBL air. Chemical tracers that are released into the study air by the launch ship make it possible to address these questions. While the major aircraft cannot spend a large fraction of their time on station seeking the center of the tracer field, it is possible to make real-time in situ measurements of a tracer like SF₆, and fine-tune the location of the sampling legs accordingly. Chemical tracers thus provide both a real-time way of keeping the observations in the right airmass and a way after the fact to confirm the extent to which the airmass dispersed over time.

Ships: At least one ship is needed to launch balloons, release tracers, measure surface water concentrations under the trajectory, and conduct longer sampling than the aircraft can. Thus, it needs to have a complete suite of aerosol and precursor measurements, including cascade impactors for determining aerosol mass and composition versus size, organic speciation, and other large-sample systems. The ship also needs to be able to follow the trajectory after balloon launch to measure the concentration of seawater species that are affecting air-sea exchange. Strong surface currents and rough sea conditions associated with the Kuroshio current in the area east of 125 E, north of 25 N may add to the challenge of following the balloon trajectory and undertaking water sampling, especially when winds oppose the current.

Aircraft: The primary aircraft (C-130) needs to have a range in excess of 2000 nmi to ensure maximum time on station as the airmass moves farther downwind; and it needs to be able to carry a payload of several tons in order to carry a complete aerosol and precursor instrument package (Table 2). Although an operations base has not yet been selected, it probably should be operated from an airport in southern Japan, to maximize access to airmasses leaving the continent. AVHRR images of springtime AOD and trajectories clearly show that the region just south of Japan is a frequent pathway for aerosols from the continent. Three Lagrangians of three 9 hour flights each would require 81 flight hours. If it were possible to do a fourth flight during the first or second Lagrangians, those hours would shorten the third one.

The CIRPAS Twin Otter would fill in most of the gaps as the primary plane refuels. Any additional aircraft could do continuous profiling as much as possible, since changes in the structure of the boundary layer are key to understanding exchanges between layers. These evolution experiments would benefit from using as many aircraft as possible.

Instrumentation: The instrumentation package for these flights is identical to that described for the survey missions. The only additions are a receiver for the location signals from the balloons, a real-time analytical system for the chemical tracer (probably SF₆), and more sophisticated communications so that communications between the planes, ships, and ops center are facilitated.

Evolution, Practical Considerations: One of the technical (or maybe political) hurdles is to measure changes where they are at their greatest. If we can find a place where concentrations change by 50%, we have the analytical sensitivity to specify some process rates well. But if we are trying to argue that a 5% change is above the noise in the aerosol property we are measuring, we will likely have difficulty proving this is significant. People will justifiably be skeptical. The largest changes, however, usually occur near sources. If military restrictions prevent us from flying or sailing within 200 nm of the Chinese coastline, for instance, some chemical systems will already have reacted so far towards completion that we won't be able to quantify the remaining changes very precisely. A major factor in the selection of an urban/industrial source region is the ability of local scientists to convince their governments of the significant gains in knowledge that could come from facilitating measurements near and perhaps even across the coastline.

In the case of dust modification, the ideal is to have airborne measurements that can study the dust as soon as it gets lofted to altitude and forms a layer that is pushed towards the sea. If one has to wait until the dust has reached the ocean to characterize the chemical and physical differences between altitudes, then we have missed observations of the most critical part of the transport story. This, however, depends on being able to fly either a foreign or domestic research aircraft within China. This aircraft would need to be equipped with the instruments to collect the most unmodified samples of dust possible. While ACE-Asia will undoubtedly make considerable

progress in addressing the program goals by working over the Pacific Ocean, the added value of measurements near aerosol sources would be considerable.

Observations of the evolution of aerosol physical and chemical properties with time are one of the most rigorous ways of constraining models that purport to represent reality. Although these experiments are complex, we have shown that they can be conducted in many parts of the world. The conditions near southern Japan appear to be ideal for the conduct of Lagrangian experiments in the springtime.

IV.E. Aerosol Interactions Research Strategy

The fundamental measurements that are needed to assess the interaction of aerosols with gas-phase chemistry involve simultaneous measurement of gas-phase and aerosol composition as a function of altitude and distance from the Asian continent. Size-specified aerosol chemical composition, to the extent possible, will be advantageous in understanding mass transport processes between the two phases. Airborne aerosol mass spectrometers will serve a key role in these measurements. These data will provide simultaneous measurements of gas-phase and aerosol-phase composition at the sea surface and aloft. While such data can quantify the amounts of species associated with the aerosol and how these amounts vary with location and type of air mass, the data will not, by themselves, reveal the mechanisms of interaction. The data will need to be evaluated with atmospheric process models that include both gas-phase chemistry and gas-to-particle conversion, as well as aerosol-phase chemistry, to attempt to constrain the most likely mechanisms of interaction. These process models will be imbedded within a three-dimensional, regional-scale tropospheric chemistry and aerosol model, which is the vehicle needed to interpret the data with respect to location and altitude and meteorological condition.

Aerosol Interactions Measurement Needs: To study the interaction of aerosols with atmospheric chemistry we will need to conduct simultaneous measurements of gas- and aerosol-phase composition at the surface and aloft, as a function of distance from the main source regions, and under different meteorological conditions. As noted earlier, these data will serve as constraints in the evaluation of three-dimensional models that describe gas- and aerosol-phase processes in the ACE-Asia region. Such models will be driven by the diagnosed meteorological fields on the days of measurement and will contain process submodels that describe the interaction of gas- and aerosol-phase chemistry. Because a three-dimensional model will be used to interpret the data, it is not necessary that the measurements be located strictly along a Lagrangian trajectory. What is required is that the data be taken in such a way as to explore the possible variation of gas and aerosol chemistry over as much of the region as can be accessed by the platforms. Thus the data should be taken at several altitudes and over a horizontal extent where effects of aerosols on gas-phase chemistry might be expected.

At least one ship will be required, on which gas and aerosol composition measurements are carried out, together with at least one aircraft. Depending on the suite of gas- and aerosol-phase measurements available on the aircraft platforms, it would be preferable to have two aircraft with complementary measurement capabilities so that the complete set of required measurements is available. The deployment strategy would be to fly the two aircraft in sufficient proximity to obtain data that would be considered to fall within a single grid cell of the three-dimensional model. A typical grid cell size would be no smaller than 10 km by 10 km, for example. The goal of the flights would be to map out a region extending as far as possible downwind of the Asian continent. At selected times, probably on the ferries out and back, the aircraft would fly over the ship at as low an altitude as possible to obtain corresponding measurements. For scientific questions G and I, at least three individual days of flights would be required, likely spread out in time over the campaign to capture different meteorological regimes to as great an extent as possible. With careful planning we might be able to use survey and evolution flights for this purpose. The measurement suite for each of scientific questions G, H, and I is given in Table 2.

IV.F. Aerosol Measurement Considerations

Aerosol inlets: The super-micron diameter aerosol will be an important component in the ACE-Asia region due to both the sea salt and mineral dust. To quantify aerosol properties in this size range will require well-characterized aerosol inlets. Participants must be prepared to characterize their inlet transmission efficiencies. This task should be clearly identified in proposals.

The difficulty of getting large particles into aircraft sampling inlets is well known (Huebert et al., 1990; Sheridan and Norton, 1998). Two developments offer the potential to improve on this during ACE-Asia: for bulk chemical sampling a new total aerosol sampler (TAS, Figure 14) collects everything that enters its tip, regardless of size. This will allow artifact-free sampling of total seasalt and dust. This device has already been flight-tested by NCAR and the University of Hawaii. A replica can be built for any of the aircraft in the experiment. For size-selective instruments, a new porous diffuser inlet (Figure 15) will decelerate air while maintaining laminar flow, thus dramatically reducing the turbulent losses of particles. Aeronautical engineers have eliminated boundary-layer

separation and turbulence in this way for years; this is its first application to aerosol inlets. Laboratory testing at Denver University has already demonstrated the concept (Seebaugh and Lafleur, 1996); flight tests of two practical devices will take place in the summer or fall of 2000. The remaining half year should be ample time to fit laminar flow diffusers to many of the participating aircraft, so that their nephelometers and sizing devices will be able to sample particles up to the 8-10 μm range.

Sampling RH: A second important consideration for intercomparability of aerosol measurements (both between platforms and between different instruments on a single platform) is sampling RH. As RH will affect size cuts and aerosol scattering, it must be sampled in the sample stream and measurement volume and recorded.

Angular scattering: One of the aerosol optical properties of specific interest is the angular scattering properties which are needed to model the aerosol albedo effect and to measure aerosol properties remotely (satellite and lidar). Although Mie theory is often used to model the aerosol phase function for complex non-spherical particles, it is not strictly valid to do this and it is not clear how much error is introduced. Due to these limitations, direct measurements of the atmospheric phase function are needed and will greatly help in understanding the complex aerosols which will be found during the ACE-Asia experiment.

For the ACE-Asia experiment we will use an innovative two wavelength polar nephelometer on the C-130 aircraft (Porter et al., 1998). The measurements will provide the aerosol phase function from 3 to 177 degrees at 1064 and 532 nm wavelengths with a sampling period of 10 sec or less depending on the aerosol concentration. Measuring the laser power and integrating over the different scattering angles, the total scattering coefficient will be obtained as well. An example of the aerosol phase function measurements is shown in Figure 16. It can be seen that for the sea spray case, the phase function has a strong forward scatter component and a rainbow oscillation near 155 degrees which is consistent with large salt aerosol. Further details are given in Porter et al., 1998.

Organics: Although the measurement of organic aerosols is a major problem, technological advances are permitting us to improve the situation: groups in Japan and elsewhere have demonstrated methods for separating organic species according to their solubility in a variety of solvents, including water of varying pH. Single particle MS methods now allow us to determine the mixing state of carbon and inorganic constituents, while HPLC now allows identification of major classes of compounds. Such primary and secondary speciation reveals much about the chemical history of the aerosol. Aerosol samples will be collected at different altitudes and ground level to achieve three objectives: (1) to measure the total organic carbon (OC) and elemental (or black) carbon (EC) content, (2) to speciate the organic aerosol at molecular levels; and (3) to determine the relative contribution of primary and secondary species to the total organic content. Characterizing the organic aerosol with every technique available (often side-by-side) will be a significant part of ACE-Asia, even though some techniques are nonspecific and others high-risk. Our ignorance of organic aerosols demands that we explore new methods.

Intercomparisons: Regularly quantifying the comparability of measurement systems on different platforms is critical. Users of the data must not be left to wonder whether the apparent difference in concentrations from two aircraft or from an aircraft and a ship or ground station are due to calibration differences or if they are due to real differences in the airmasses sampled. Even after instruments have been extensively intercompared, new leaks, electrical interference, drift in calibration standards and other realities of field science can introduce doubts about the reality of small differences. Wingtip to wingtip flights are the only way to demonstrate that on that day, under those conditions, both planes were measuring the same thing. Planes (and ships and ground stations) working together must include comparison legs.

The same is true of comparisons between ships and aircraft. It is not a trivial matter for platforms moving 100 m/s and 5 m/s to sample the same air, but it can be done. As in the Cape Grim intercomparisons in ACE-1, a plane can fly a half-hour leg upstream of a ship or ground station (slightly offset so that the ship does not see aircraft exhaust), ending with a flyby as close as possible. Then the ship or ground station can time its sampling with the wind speed so that it samples (over time) the same air that the plane sampled (over space). This strategy can be modified to suit short or long sampling intervals, but most of the relevant measurement systems can be compared.

Complementary measurements: Another compelling reason to sample the same air is that the suite of measurement systems will no doubt be different on the various platforms: the extra information that can be derived by measuring all those species with all those instruments at the same time is of tremendous value. An example might be that a plane with extensive aerosol measurements (the C-130) but without an OH instrument could sample on a coincident leg with a plane having OH measurements (the NASA DC-8), thus expanding the suite of values available to both sets of investigators. Common legs flown by multiple aircraft can constrain models of photochemistry, aerosols, and transport better than either plane could alone.

IV.G. Meteorological Overview

Selection of an optimum time for the AA-SEC intensive depends upon the typical transport patterns. In

some arid areas of China mineral aerosol is raised from the surface all year around, but occurrence of offshore transport increases from late winter to a maximum in April, followed by a gradual decline into summer. Transport patterns are dominated by midlatitude storms, and vary daily and on an interannual scale. Conditions are favorable for transport out over the ocean in the middle and upper troposphere for much of the year. However, at and near the surface, outflow conditions in the winter and early spring gradually give way to inflow. The northern edge of the onshore flow progresses from 20°N to 35°N and the depth of the warm-air layer increases through April into May. The frequency and intensity of precipitation increase markedly with the onset of inflow. Given these factors and recognizing inescapable variability, we selected approximately March 11 - April 15 as the proposed window of operations.

A basic picture of the meteorological conditions in Asia and the western North Pacific can be obtained from the results and discussion of earlier atmospheric chemistry research programs and by examination of relevant meteorological data and model simulations. In this brief overview we use both of these approaches, drawing mostly on major research campaigns and on in situ observations. The transport environment is influenced by forcings and variations over a very wide area and over a range of time scales, but we emphasize here the circulation over and east of China, between 20 and 50°N, and focus the discussion on the late winter/early spring period. During the winter months deep layers of cold air present over Siberia result in exceptionally high surface pressures there; the surface anticyclone present at that time contrasts with a weaker cyclonic circulation during the summer months. This contrast parallels the variation in offshore-onshore flow much further to the east and south, at the coast and out over the ocean: outflow conditions are typically present in the winter and spring, while onshore winds (inflow conditions) prevail during the summer and fall. The anticyclone weakens but the surface air remains cold in NE Asia and during the spring months cyclonic disturbances initiated in or propagating through the Siberian plains push surface cold fronts through the mountainous terrain south and west of Lake Baikal, the Altai-Sayan and Tianshan ranges which lie just northwest and farther west of the Mongolian plain, respectively. A springtime maximum in storm formation is present downstream of these mountains, with the dynamics driven by lee cyclogenesis and the presence of these surface thermal contrasts (Chen et al., 1991). The strong surface winds and uplifting associated with the cold fronts accompanying these secondary cyclones are responsible for the well-known springtime maximum in dust raised from the surface in eastern Asia (e.g., Middleton et al. 1986; Merrill et al. 1989, Xu and Hu, 1996). It has been hypothesized that the dust can itself strengthen the frontal temperature contrast (Chen et al., 1995). In any event the dust storms have a critical role in the aerosol environment (e.g., Zaizen et al. 1995, Zhou et al., 1996, Zhang et al., 1998). Also, the turbulent winds, low-level uplift and strong upper-level winds present in these systems enhance the offshore flow of pollutants as well. It is important to note, however, that the characterization by the PEM-West B science team that the flow was offshore during February-March was based on a selective sampling protocol, designed to investigate the impact of natural and human activities on the chemistry of the northwestern Pacific area. That is, periods of strong offshore flow in the near-surface layers alternate with typically weaker onshore flow, while at higher elevations the component of the flow from the west increases throughout the troposphere. The pattern of a cyclonic flow centered in the northern part of this region and an anticyclonic shear axis to the south prevails, but the locations of the centers and consequently the extent of these areas varies. The latitude of the shear axis on the northern side of the anticyclonic flow also varies, from 20°N to 30°N.

The conditions which are most favorable for Lagrangian experiments exist in the anticyclonic flow, and are most likely to be accessible from southern Japan when the shear axis is further to the north. Thus the monthly mean velocity field below 700 hPa (~ 3km) can exhibit flow parallel to the coast or even onshore during spring months in some years. However, typical conditions have the flow offshore often and in the mean, as illustrated in Fig. 17. The influence of the anticyclonic flow is evident south of 40°N and the more westerly flow is obvious north of 40°N. A similar pattern was present during PEM-West B (Merrill et al., 1997; Fig. 1c). Composition measurements indicated the importance of continental outflow very directly, both in aggregate (Talbot et al., 1997) and in consideration of data from vertical profiles alone (Gregory et al., 1997).

The middle and upper tropospheric winds are westerly throughout this area, and in fact the peak jet stream-level winds south of Japan are the strongest in the world, exceeding 60 m s⁻¹. The contrast between cold air over the continent and warm air over the ocean sustains these high winds, and even as the maximum speed decreases from February through May the occurrence of midlatitude storms remains high. There is a gradual shift, however, with both the genesis and passage of storms becoming less common in the southern area of this region as the cold air retreats to the north through March into April. Cloudiness is common throughout this period in this area, with mean cloud amounts exceeding 65% over the ocean areas south of Japan, and with low clouds approximately 50% of this total. However, as illustrated in the satellite images in the examples discussed below cloud amounts are lower in the anticyclonic conditions most favorable for Lagrangian experiments. The climatological precipitation distribution for eastern China and the area of Korea and Japan is easily understood in the context of the winter offshore flow and the

summer onshore flow: precipitation amounts in the coastal areas increase with the arrival of the warm, onshore flow in the southern-most areas in early spring, and this pattern moves north as the onshore flow extends to higher latitudes. Thus on average areas of southern Japan go from <100 mm of precipitation in February to > 200 mm in April; correspondingly areas in southern Korea go from 25-50 mm in February to 75-100 mm totals in April. A secondary maximum in the frequency of cyclone passage and cyclogenesis in the spring months occurs between the south coast of Japan and 30 N east of 125 E, influenced in part by the path of the Kuroshio current.

Examples of upper-level transport patterns during outflow conditions are included in the PEM-West B meteorological overview (Merrill et al., 1997) and in other papers in the PEM-West collection. Analysis of isentropic trajectories at free-tropospheric levels for more recent years (1995-1998) (not shown) exhibit significant interannual and day to day variability in the specific features (the latitude of the transition to anticyclonic flow, for example). A statistical analysis of trajectories calculated within the boundary layer (employing the Hysplit model, which is more appropriate there than the isentropic model), selected to satisfy "attainable" Lagrangian experiment conditions using a base of operations at Fukuoka, Japan, is available at <http://www.aero.obs-mip.fr/ace/ace3/trajs/ver2.html>. The results indicate that the interannual variability always encompasses a satisfactory likelihood of achievable Lagrangian situations. Also available is a case study, again using Hysplit trajectories, illustrating the types of meteorological products needed for operational and post-mission analysis; these include satellite cloud images critical to the assessment of potential vertical exchange (<http://www.aero.obs-mip.fr/ace/ace3/trajs/case1.html>).

V. Project Management Structure

V.A. ACE-Asia Components

The International Global Atmospheric Chemistry Program (IGAC) has conducted three major experiments to characterize aerosols, their effects, and the processes that control their distribution and composition. ACE-1 (the first Aerosol Characterization Experiment) studied natural, unpolluted aerosols in the vicinity of Tasmania in 1995 (Bates et al., 1998). TARFOX (the Tropospheric Aerosol Radiative Forcing Experiment) focused on the radiative impact of anthropogenic aerosols off the Atlantic coast of North America (Russell et al., 1999). The ACE-2 experiment was directed at European pollution aerosols and their impact on the Atlantic.

From a management and planning perspective, the ACE-Asia Science Team has recognized that the overall ACE-Asia experiment needs to be broken down into four components. Each of these components is a logical unit that shares many overlapping scientific objectives with the others but that has some unique strategic approaches to addressing certain of these objectives:

- A. Network Operations, 2000-2004
- B. Survey and Evolution Intensive, March/April 2001, AA-SEC
- C. Indirect Forcing Intensive, Spring 2002 or 2003
- D. Direct Forcing Intensive, Spring 2003 or 2004

Each of these components will have its own science and implementation plan that describes its specific scientific questions and proposes strategies for answering them.

V.B. AA-SEC Management Structure

- 1) The Science Team includes all PIs participating in the project from every country.
- 2) The ACE-Asia Survey and Evolution Component (SEC) will be lead by a Scientific Steering Committee (SSC), chaired by the ACE-Asia Lead Scientist, Barry Huebert. The SSC will consist of:
 - a) the convenors (or a designated Activity Committee member) of ACAPS, APARE, MAGE, and GIM,
 - b) the chair of each ACE-Asia National Committee,
 - c) a representative of each major observing facility and platform, including the most comprehensive surface sites.

V.C. Interaction with other programs

ACE-SEC plans to work closely with a NASA GTE experiment, TRACE-P (Transport and Chemical Evolution over the Pacific), that is planning to study Asian outflow to the Pacific in March and April of 2001. Although plans are not yet finalized, it will probably involve flights of both NASA's P-3 and DC-8 aircraft. The focus of TRACE-P is on photochemistry in Asian outflow, which is complementary to the ACE-Asia aerosol-oriented focus. Based on the interactions so far between the two science teams and their common parentage by the IGAC APARE activity, we expect to coordinate some part of our flight hours and ship observations to take advantage of the complimentary sets of instruments brought by the two programs. We also hope to collaborate with the PICASSO-CENA (lidar in space) science team, perhaps to collect in situ data as they fly an airborne simulator.

V.D. World Wide Web (Participants and Platforms)

Further information can be found on the ACE-Asia web site: <http://saga.pmel.noaa.gov/aceasia/>. A list of all persons having asked to be informed about ACE- Asia is available at the web site. Potential ACE-Asia platforms and the instrument working groups are also listed. The web page version of the ACE-SEC Science and Implementation Plan will be updated roughly every six months after international science team meetings.

VI. Data Management Plan

Data Archive: The development and maintenance of a comprehensive and accurate data archive is a critical step in meeting the goals of the ACE. The overall ACE data management philosophy is to make the completed data set available to the world research community as soon as possible in order to better incorporate aerosols into global climate models. A centralized data archive will be established to combine the entire ACE-Asia data set. This integrated data base will allow users a single access to the variety of measured and derived fields obtained during ACE-Asia. A central data archive is sometimes a difficult issue, since many groups and nations have traditionally kept their data to themselves. However, the benefits far outweigh the liabilities as everyone has access to a much larger data set than they could possibly obtain or pay for alone.

The following data protocols, established for ACE 1 and 2, will hold for all ACE-Asia participants:

1. All investigators participating in ACE-Asia must agree to promptly submit their data to the central data base to facilitate intercomparison of results, quality control checks and inter-calibrations, and an integrated interpretation of the combined data set.
2. All data shall be promptly provided to other ACE investigators upon request. A list of ACE-Asia investigators will be maintained by the SSC and will include the principle investigators directly participating in the field experiment and the modellers who have provided guidance in the planning of ACE Asia activities.
3. During the initial data analysis period (one year after the data were collected), no data may be provided to a third party (journal articles, presentations, research proposals, other investigators) without the consent of the investigator who collected the data. This initial analysis period is designed to provide an opportunity to quality control the combined data set.
4. It is the intent of the ACE science team that all data will be considered public domain at the end of the ACE-Asia field experiment and that any use of the data will include either acknowledgment or co-authorship at the discretion of the investigator who collected the data.

VII. Participating Scientists

The following have either submitted text, attended a planning meeting, or made a definite commitment to participate. Without question there are many inadvertent omissions. (* = contributed to S&IP)

China

Qian Fenlan (NRCMEF)
Huanlin Hu (Anhui Inst. of Optics and Fine Mechanics, AIOFM)
Darren Lu (IAP)
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Jinhuan Qiu (IAP)
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IX. Tables

Table 1. Organic Constituents of Atmospheric Particulate Matter

Classes of organic compounds that have been measured in atmospheric particles or are predicted to be present in atmospheric particles based on thermodynamic arguments and measurements in other media (Rogge et al., 1993; Saxena and Hildemann, 1996).

<i>n</i> -alkanes	Aliphatic (di)carboxylic acids
<i>n</i> -alkanoic acids	Aromatic (di)carboxylic acids
Aldehydes	Glyoxal
Esters	Organic peroxides
Diterpenoid acids	Ketoacids
Aromatic polycarboxylic acids	Polyols
Polycyclic aromatic hydrocarbons	Hydroxyamines
Polycyclic aromatic ketones	Amino acids
Polycyclic aromatic quinones	Nitrophenol

Table 2. Measurement Priorities**Priority (1 – essential, 2 – value added, 3 – complementary, 0 – not needed)**

The Scientific Questions are repeated here for readability with the Table.

Aerosol Survey

What are the vertical and regional distributions of aerosol concentrations and properties under various meteorological conditions?

- A. What are the relative contributions of sulfate, nitrate, carbonaceous material, sea salt, primary anthropogenic particles, and dust to the radiative properties (absorption, total and angular scattering) of aerosols?
- B. How do the interactions between these species (their state of mixing) affect their radiative properties?
- C. How are the distributions affected by specific meteorological events?

Aerosol Evolution

How do aerosol properties evolve as they move offshore?

- D. How do aerosol properties and process rates evolve with time (distance) over the ocean?
- E. What are the lifetimes or removal rates of particles?
- F. What is the rate of exchange of particles and precursor gases between the free troposphere, buffer layer, and boundary layer?

Aerosol Interactions with Gas-Phase Atmospheric Chemistry

What role does the atmospheric chemistry of the Asian-Pacific region play in influencing aerosol properties and evolution, and likewise, how does the Asian aerosol affect tropospheric chemistry in the region?

- G. How does mineral dust interact with atmospheric gas-phase chemistry?
- H. How does sea salt interact with gas-phase chemistry?
- I. How does the anthropogenic/urban aerosol interact with gas-phase chemistry?

Measurement	Platform				Scientific Question								
	C-130	Twin Otter	Ship	Land	A	B	C	D	E	F	G	H	I
<i>H₂SO₄</i>	2	2	3	3	0	0	0	2	2	2	0	0	0
<i>OH</i>	2	0	3	3	0	0	0	2	2	2	0	0	0
<i>NMHCs</i>	2	0	2	2	0	0	0	2	0	2	0	0	0
<i>DMS</i>	2	0	1	3	0	0	0	1	0	2	0	0	0
<i>SO₂</i>	1	1	1	1	0	0	0	1	1	2	1	1	1
<i>NH₃</i>	2	2	1	3	0	0	0	2	2	3	0	1	0
<i>NO_x</i>	2	0	1	2	0	0	0	2	3	3	0	1	1
<i>NO_y</i>	2	0	1	2	0	0	0	2	3	3	0	1	1
<i>HNO₃</i>	2	0	1	2	0	0	0	2	3	3	2	1	1
<i>O₃</i>	1	3	1	1	0	0	0	2	3	2	1	1	1
<i>CO</i>	2	0	2	2	0	0	0	2	3	2	0	0	0
<i>H₂O₂</i>	2	0	2	2	0	0	0	2	2	2	2	0	0
<i>HCl</i>	2	0	1	3	0	0	0	2	2	3	2	1	0
<i>Cl₂</i>	2	0	1	3	0	0	0	2	2	3	2	1	0
<i>Br₂</i>	2	0	1	3	0	0	0	2	2	3	2	1	0
<i>HBr</i>	2	0	1	3	0	0	0	2	2	3	2	1	0
<i>H₂O</i>	1	1	1	1	1	1	1	1	1	1	1	1	1
<i>Rn-222</i>	0	0	1	1	2	2	1	3	3	3	1	3	1
<i>Precipitation</i>	3	0	1	1	3	3	1	3	1	0	0	0	0
<i>CCN</i>	3	3	3	3	3	3	3	3	3	0	0	0	0
<i>Cloudwater chem.</i>	2	0	0	0	0	0	0	2	2	0	2	2	2
<i>Single-particle MS</i>	1	1	1	1	1	1	1	2	2	0	1	1	1
<i>Single-Particle Electron Microscopy</i>	1	0	1	1	1	1	1	2	2	2	1	1	1

Measurement	Platform				Scientific Question								
	C-130	Twin Otter	Ship	Land	A	B	C	D	E	F	G	H	I
<i>Mineralogical comp.</i>	3	0	2	1	1	1	1	3	0	0	1	2	2
<i>Size-resolved ions</i>	1	1	1	1	1	1	1	3	1	0	1	1	1
<i>Size-resolved trace element</i>	2	1	1	1	1	1	1	3	3	0	1	0	1
<i>Sub-10nm comp.</i>	3	0	0	0	3	3	3	3	0	0	0	0	0
<i>Real time SO₄/NO₃/OC/EC</i>	2	2	1	1	1	1	1	2	0	0	2	0	2
<i>Real time Na</i>	1	0	1	0	1	1	1	2	0	0	0	1	0
<i>Speciated organics</i>	0	1	1	1	1	2	1	1	3	0	2	0	2
<i>Size-resolved organics (EC/OC)</i>	0	1	1	1	1	1	1	2	2	0	2	0	2
<i>Aerosol phys. Size distrib. UF mode</i>	2	1	3	2	3	3	1	1	3	2	3	3	3
<i>Aerosol phys. Size distrib. Aitken mode</i>	1	1	1	1	1	1	1	1	1	1	1	1	1
<i>Aerosol phys. Size distrib. Accum. mode</i>	1	1	1	1	1	1	1	1	1	1	1	1	1
<i>Aerosol phys. Size distrib. Coarse mode</i>	1	1	1	1	1	1	1	1	1	1	1	1	1
<i>Total mass, ions TAS</i>	1	3	1	1	1	1	1	2	0	0	0	0	0
<i>Hygroscopic growth</i>	2	1	1	1	1	1	1	2	0	0	1	0	1
<i>Aerosol phase function, polar neph</i>	1	0	0	2	1	1	0	1	0	0	0	0	0
<i>Nephelometer</i>	1	1	1	1	1	1	1	2	3	0	1	0	0
<i>Absorp.</i>	1	2	1	1	1	1	1	2	3	0	1	0	0
<i>Radiative fluxes</i>	2	2	1	1	1	1	1	2	3	0	0	0	0
<i>LIDAR – aerosol backscat.</i>	1	0	1	1	1	2	1	1	2	1	0	0	0
<i>Fluxes *</i>	1	1	0	0	0	0	1	1	1	1	0	0	0

*Eddy correlation - O₃, momentum, heat, water vapor, DMS as a tracer, SO₂ flux to surface

Table 3. C-130 Recommended Payload

Highest Priority - Essential

- A. Aerosol size, chemistry, and optical properties
 - Impactor – bakeable for organics & EC/OC (High-flow Moudi?)☹
 - Impactor – ions and mineral aerosols (High-flow Moudi?)☹
 - Total Aerosol Sampler, TAS – bakeable for organics & EC/OC
 - TAS – ions and minerals
 - Real-time Na analyzer*☹
 - Single-particle Mass Spec*☹
 - Single-particle Electron Microscopy collections☹
 - Aerodynamic Particle Sizer☹
 - SMPS
 - FSSP
 - PCASP☹
 - Absorption Instrument/aethalometer☹
 - Wet & Dry Nephelometers☹
 - 180° Nephelometer for lidar ratio☹
 - Polar Nephelometer (external, so no inlet)*
 - Total Particle Number (CN) ☹
- B. Radiation/remote sensing Package
 - Shadowband Radiometer*
 - Tracking Sun Photometer (NASA Ames)*
 - UV flux
 - Lidar
- C. Gases, precursors, and photochemistry
 - Ozone
 - H₂O
 - SO₂
- D. Turbulent fluxes of Sensible Heat, Momentum, Moisture, Ozone, SO₂, and DMS
- E. Lagrangian Support
 - Balloon Tracking Receiver
 - Real-time SF₆ Analyzer
- F. RAF Standard state variables, winds, location, etc.

Second Priority

- Nanoparticles☹
- Fast sulfate & nitrate☹
- Liquid cloudwater chemistry
- Cloud Condensation Nuclei☹
- DMS
- H₂O₂ **
- CO (RAF)
- CO₂ (RAF)
- NO/NO₂/NO_y
- HNO₃
- NMHCs & CFCs (Tai Chen, Taiwan)
- OH**
- NH₃
- H₂SO₄ vapor**
- MSA vapor**

*High priority but not a show-stopper

☹ These instruments would get their air samples from a few non-turbulent porous-diffuser inlets.

** These measurements may only be available through coordination with TRACE-P.

Table 4. Twin Otter Recommended Payload

Existing Instrumentation	Parameter measured
A. Aerosol size, chemistry, and optical properties	
Automated Classified Aerosol Detector – ACAD	Particle Size Distribution; 0.003 μm <Dp<0.2 μm (10 s)
PCASP	Particle size distribution; 0.1 μm <Dp<3 μm
Cloud, Aerosol and Precipitation Spectrometer	Particle Size Distribution; 0.3 μm <Dp<30 μm
Optical Array Probe-230	Particle Size Distribution; 30 μm <Dp<600 μm
Cloud Condensation Nuclei Instrument	Cloud Condensation Nuclei; 0.1%<Sc<2.0%
Ultra-fine Condensation Particle Counter	Particle concentration for Dp>0.003 μm
Nephelometer, 3 color	Integral Scatter at 0.45, 0.55, and 0.7 μm
Humidigraph	Integral Scatter at 0.545 μm at high and low RH
Soot Photometer	Differential transmissivity of a sampling filter
Filter Sampling System	Aerosol Mass and Composition (organics)
Aerosol Mass Spectrometer	Composition of aerosol particles; 40 μm <Dp<2.5 μm
High Resolution Water Vapor Probe	Water vapor concentration
Cascade Impactor(MOUDI)	Size classified particle composition(5 stages<2.5 μm , high flow)
B. Radiation	
Pyranometer (2 units)	Total solar flux-Direct & scattered for 0.285 μm <l<2.8 μm
Pyranometer	Partial solar flux-Direct and scattered for 0.715 μm <l<2.8 μm
Pyrgeometer	Total infrared flux; l>4 μm
Total Ultraviolet Radiometer	Total ultraviolet flux; 0.295 μm <l<0.385 μm
Tracking Sun Photometer (NASA Ames)	Aerosol Optical Depth
C. Meteorological and Flight Parameters	
Temperature	
Dew Point Temperature	
Static Pressure	
Wind, 5 hole probe	
Turbulence, BAT-System	
TANS Vector	
NovAtel GPS Differential	
Under Development	
Atmospheric Pressure Chemical Ionization Mass Spectrometer	DMS, MSA, SO ₂ , NH ₃

Note: The Twin Otter investigators welcome proposals for other instrumentation of appropriate size and power consumption to be installed on the Twin Otter for ACE-Asia. Interested investigators should contact Prof. John Seinfeld at Caltech.

Table 5. NOAA-UNOLS Ship Recommended Measurements

- A. In-situ aerosol measurements:
 - 1. Chemical:
 - a) Size resolved chemical sampling for inorganic ions, mineral dust, and total and organic carbon. (7 stage Berner-type impactors, PMEL)
 - b) Single particle analysis by mass spectrometry
 - c) Single particle analysis by electron microscopy (PMEL)
 - d) Higher time resolution (1-2 hour) measurements of total sulfur, carbon, nitrogen using a two stage impactor thermal technique (PMEL?)
 - e) Sodium particle number size distribution
 - f) Size resolved sampling for organic speciation
 - 2. Physical and optical:
 - a) Number size distribution from 5 to 10,000 nm diameter using tandem DMPS and APS system (PMEL)
 - b) Size resolved mass (gravimetric) size distribution (7 stage Berner-type impactors, PMEL).
 - c) Total particle number (PMEL)
 - d) Total and sub-micron light scattering and backscattering by aerosols at 3 wavelengths (PMEL)
 - e) Total and sub-micron light absorption by aerosols (PMEL)
 - f) Aerosol hygroscopic growth (H-TDMA)
 - g) Satellite receiving station to obtain real-time images of aerosol backscatter
- B. Column measurements:
 - 1. Aerosol optical depth using hand held sunphotometers (PMEL)
 - 2. LIDAR measurements of aerosol vertical distribution
 - 3. 180° backscatter (for lidar interpretation)
- C. Trace gases
 - 1. DMS (PMEL)
 - 2. SO₂
 - 3. Ozone (PMEL)
 - 4. NMHC
 - 5. Reactive halogens
 - 6. Nitrogen oxides
 - 7. Radon (PMEL)
 - 8. CO
 - 9. Ammonia
 - 10. Hydrogen peroxide
- D. Seawater measurements
 - 1. DMS (PMEL)
 - 2. Iron
 - 3. Ammonia and pH
 - 4. SST, salinity, nitrate, chlorophyll (PMEL)
- E. Meteorological measurements
 - 1. Surface meteorological data (PMEL)
 - 2. Integrated sounding system (ISS) balloon, profiler, RASS (NCAR)
- F. Lagrangian support
 - 1. Smart balloons
 - 2. PFC tracer

Table 6. AA-SEC Enhanced Surface Site Measurements

As part of the AA-SEC intensive we propose to equipment one “super-site” with the following instrumentation. The location of this “super-site” will most likely be Cheju Island, in the center of the AA-SEC study area. Some of these measurements will already be located at this site as part of the AA-Network component, the TRACE-P study, and ongoing activities of Korean scientists. We will duplicate many of the aerosol measurements at a site near the mineral dust source region.

- A. In-situ aerosol measurements:
 - 1. Chemical:
 - a) Size resolved chemical sampling for inorganic ions, mineral dust, and total and organic carbon.
 - b) Single particle analysis by mass spectrometry
 - c) Single particle analysis by electron microscopy
 - d) Higher time resolution (1-2 hour) measurements of total sulfur, carbon, nitrogen using a two stage impactor thermal technique
 - e) Size resolved sampling for organic speciation and water solubility
 - f) Mineral composition to distinguish reacted from unreacted mineral aerosol as well as identify source regions
 - g) Precipitation chemistry
 - 2. Physical and optical:
 - a) Number size distribution from 5 to 10,000 nm diameter using tandem DMPS and APS system
 - b) Size resolved mass (gravimetric) size distribution.
 - c) Total particle number
 - d) Total and sub-micron light scattering and backscattering by aerosols at 3 wavelengths
 - e) Total and sub-micron light absorption by aerosols
 - f) Aerosol hygroscopic growth (H-TDMA)
 - g) Scattering and backscattering as a function of RH ($f(RH)$)
 - h) Aerosol phase function measurements (from 3-177 degrees) at two or more wavelengths
- B. Column measurements:
 - 1. Aerosol optical depth using hand held sunphotometers
 - 2. LIDAR measurements of aerosol vertical distribution
 - 3. 180° backscatter (for lidar interpretation)
- C. Trace gases
 - 1. SO₂
 - 2. Ozone
 - 3. NMHC
 - 4. Nitrogen oxides & HNO₃
 - 5. CO
 - 6. H₂O₂
- D. Meteorological measurements
 - 1. Surface meteorological data (PMEL)
 - 2. Integrated sounding system (ISS) balloon, profiler, RASS (NCAR)
- E. Lagrangian support
 - 1. Smart balloons
 - 2. PFC tracer

Table 7. Platform Resources Needed

NOAA or UNOLS Ship	35 ship-days to and from a Japanese port (near the Ops Center), plus transit time
NCAR C-130	72 hours survey flights (8 flights @ 9 hours) 81 hours Lagrangian flights (9 flights @ 9hrs) Ferry to and from US: most direct route, but most instruments will be operated enroute
CIRPAS Twin Otter	100 research hours (15 flights of 6-8 hours) No science will be done during ferry

Table 8. Tentative Schedule

Nov 1997	Science Team Meeting (Nagoya, Japan) http://saga.pmel.noaa.gov/aceasia/nagoya_report.html
Nov 1998	Science Team Meeting (Cheju Island, Korea) http://saga.pmel.noaa.gov/aceasia/cheju98.html
Mar 1999	AA-SEC workshop, Cal Tech
Jun-Dec 1999	Instrumentation intercomparison workshops
Nov 1999	Science Team Meeting (China) Station Network site visits
Jan 2000-Dec 2004	Surface Network Operations
Spring 2000	SSC meeting
Fall 2000	Science Team Meeting near Operations Center Site
Mar-Apr 2001	Intensive Observation Period AA-SEC
Fall 2001	Data Workshop/Science Team Meeting
Spring 2002	SSC meeting
Fall 2002	Data Workshop/Science Team Meeting
March/April 2003	Second Intensive Observation Period Cloud-Aerosol Interaction Study ? Direct Radiative Forcing Study ?
Fall 2003	Data Workshop/Science Team Meeting
Jan 2002-2006	Data Archiving, Meeting Presentation, Journal Special Issues

X. Figures

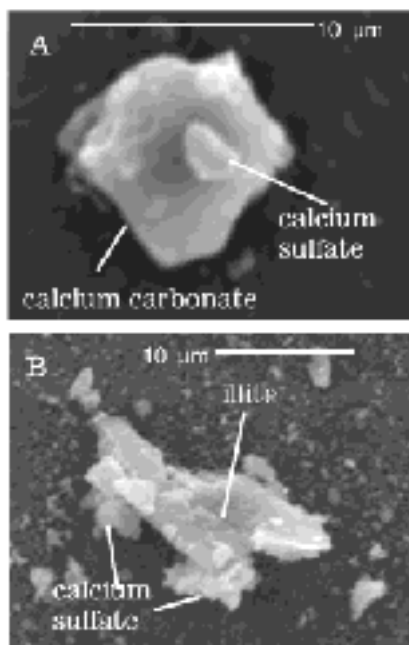


Figure 1. Secondary electron images of aerosol particles from (A) Shapoutou, China (in the loess source area) and (B) Qingdao, at the Chinese coast. In image (A) is a large calcite particle, probably of natural soil origin, with a small particle of calcium sulfate, indicating very limited reaction of the carbonate with SO_2 . In image (B) is a grain of illite, a common clay in loess, aggregated with small crystals of calcium sulfate. This particle probably was originally a natural, soil-derived aggregate of illite and calcite that underwent complete reaction of the carbonate with added sulfuric acid during transport. Images from J. Anderson

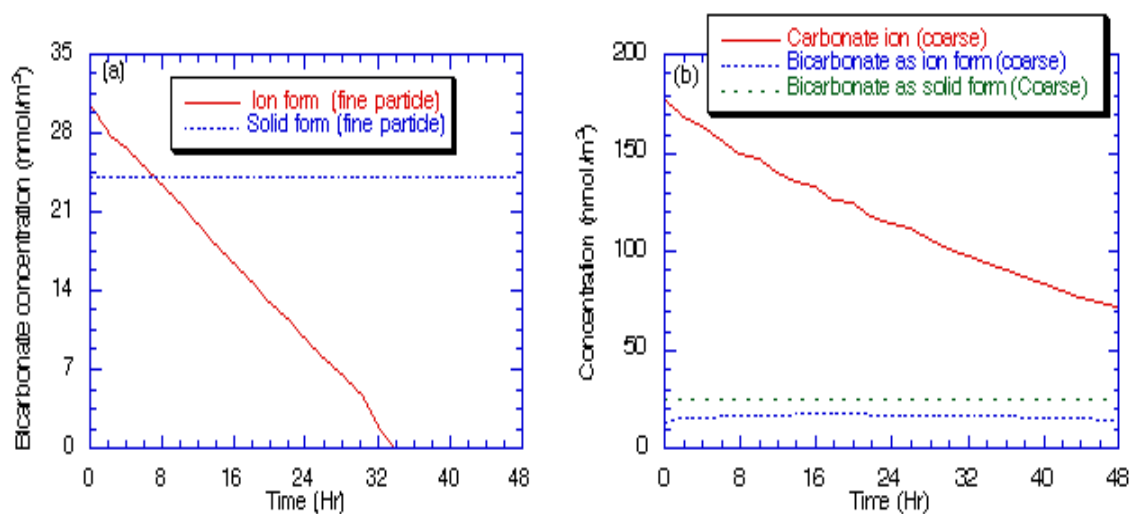


Fig. 2. Aerosol carbonate and bicarbonate vs time, showing the impact of particle size on reaction rate. G Carmichael.

West Pacific (1500 km east of Sapporo, Japan)

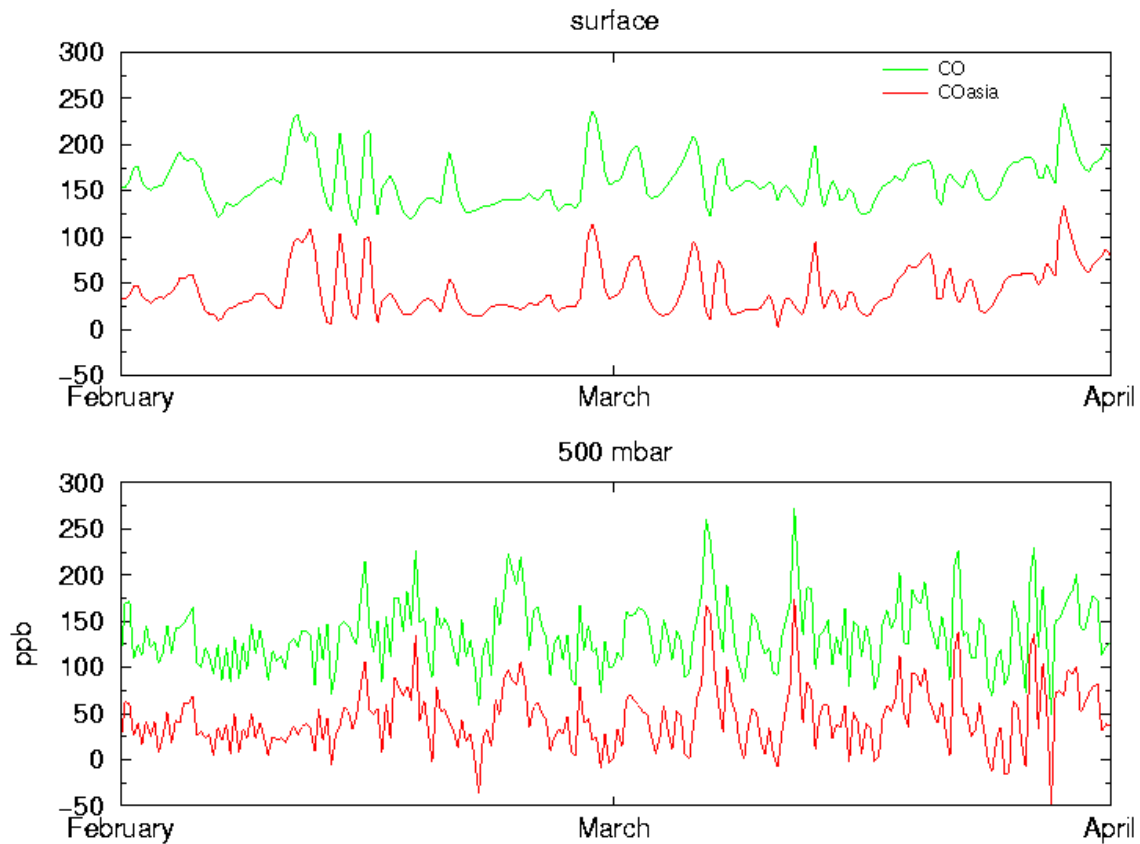


Figure 3. CO mixing ratios measured 1500 km downwind of Japan. The upper figure (a) shows data from the surface while the lower figure (b) shows measurements at 500 mb. The values are the "Asia contribution to CO" which is calculated as the difference between a base simulation and a simulation without Asian CO emissions. G. Carmichael, http://www.cgrer.uiowa.edu/people/mphadnis/ace_asia.html

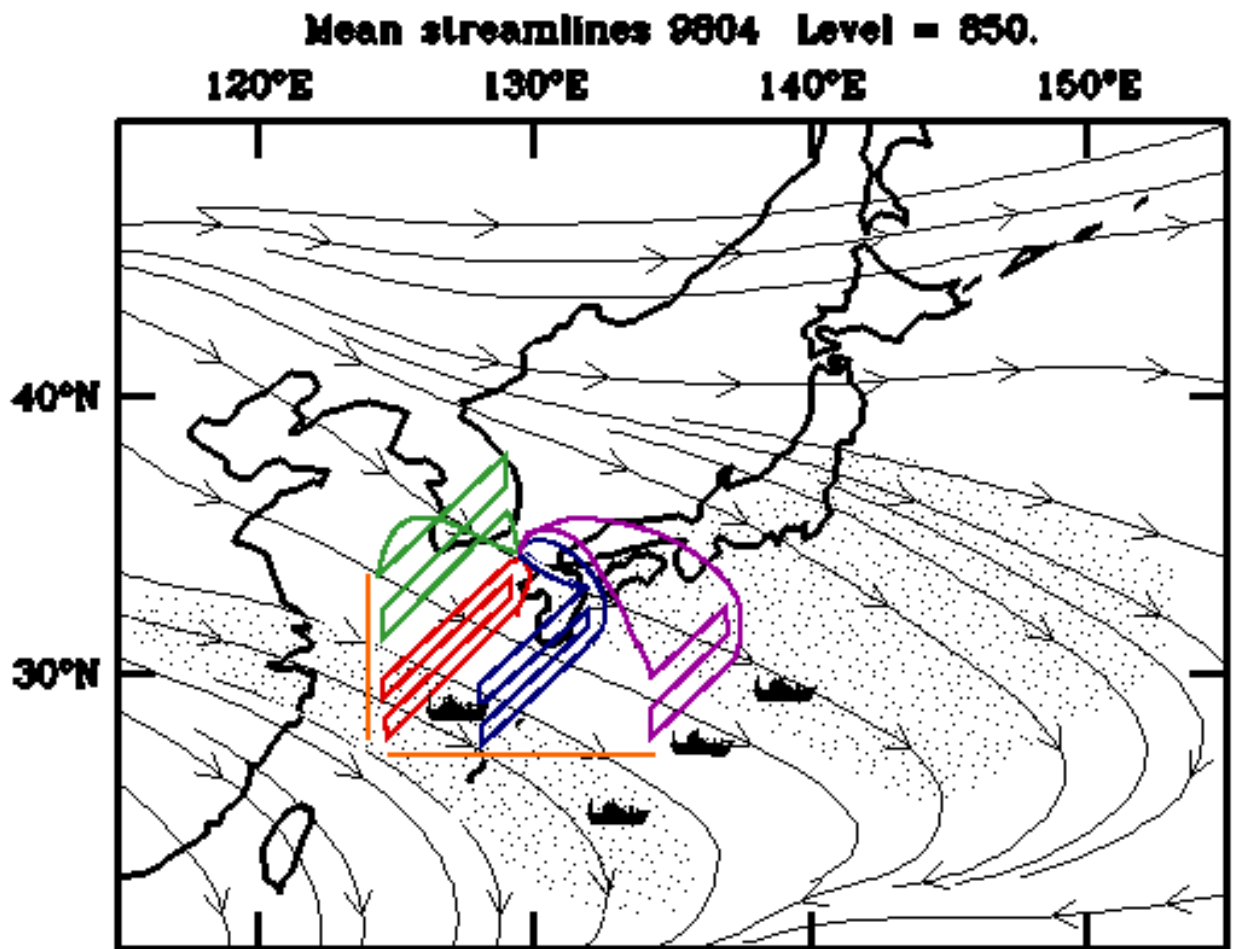


Figure 4. Four possible wall pattern flights. The green legs represent a higher-altitude stack flown above that of the red stack. Walls can be configured as short legs at many levels or longer legs (crossing several features or plumes) at a few levels, depending on the complexity of the vertical structure as determined by lidar. Ships would be used to measure time-series and collect samples that require multi-hour sampling times, such as for cascade impactors and organic speciation. Multiple ships would enable sampling within and outside of plumes. The NOAA ship (at least) needs to also have a UCAR Integrated Sounding System, that will give a time history of changes in inversion heights and thermodynamic variables.

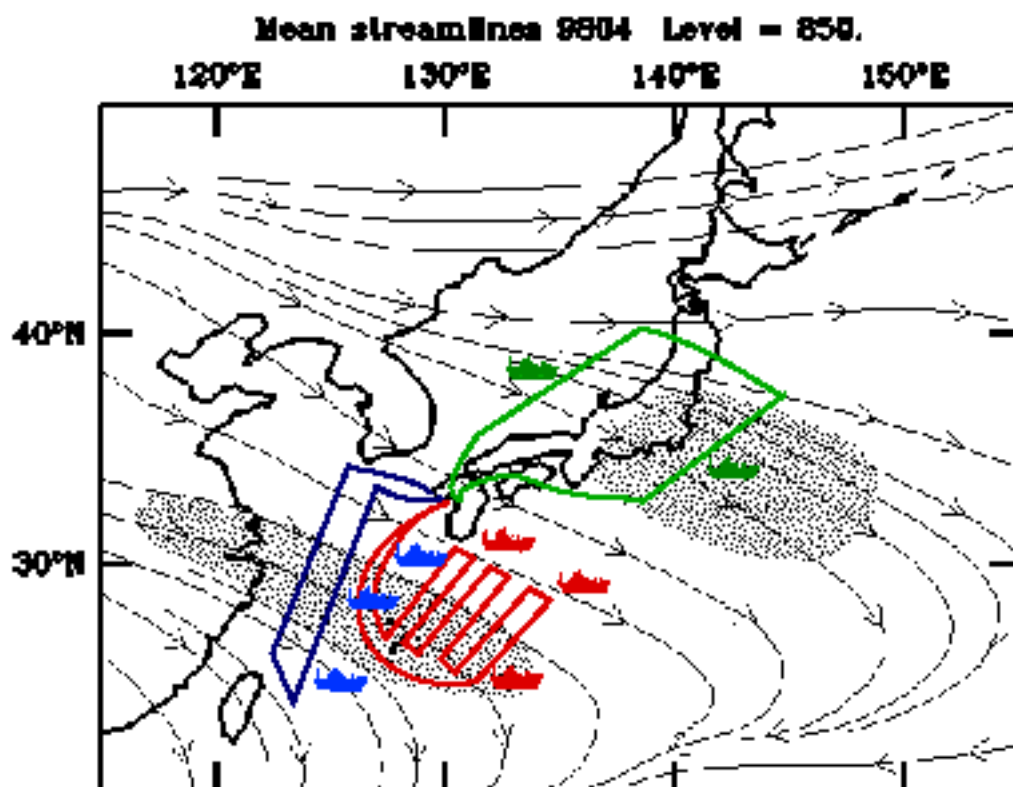


Figure 5. Three different types of horizontal survey operations. These could be chosen to fly across interfaces between a plume and less impacted air or entirely within one type of air. The blue flight path and ships describe a long transit at one altitude, designed to span large features. The red operation would examine the interface much more intensely, with multiple crossings between the two regimes. The green survey is essentially a perturbation experiment, looking at the influence of sources by sampling upwind and downwind. Based on satellite images and the resolution of candidate models, a choice would be made between long legs at just a few altitudes or shorter legs at many altitudes. These choices would largely be made in consultation with modelers and satellite images prior to the flight. It would be possible to do any of these with a single ship, but it is clearly desirable to have surface measurements both within and out of a plume, for instance, which would require two or more ships. Two or more aircraft would likewise allow for surveying a broader region at one altitude or doing multiple altitudes in the same spatial domain.

ACE-Asia Sea Salt Gradient Flight

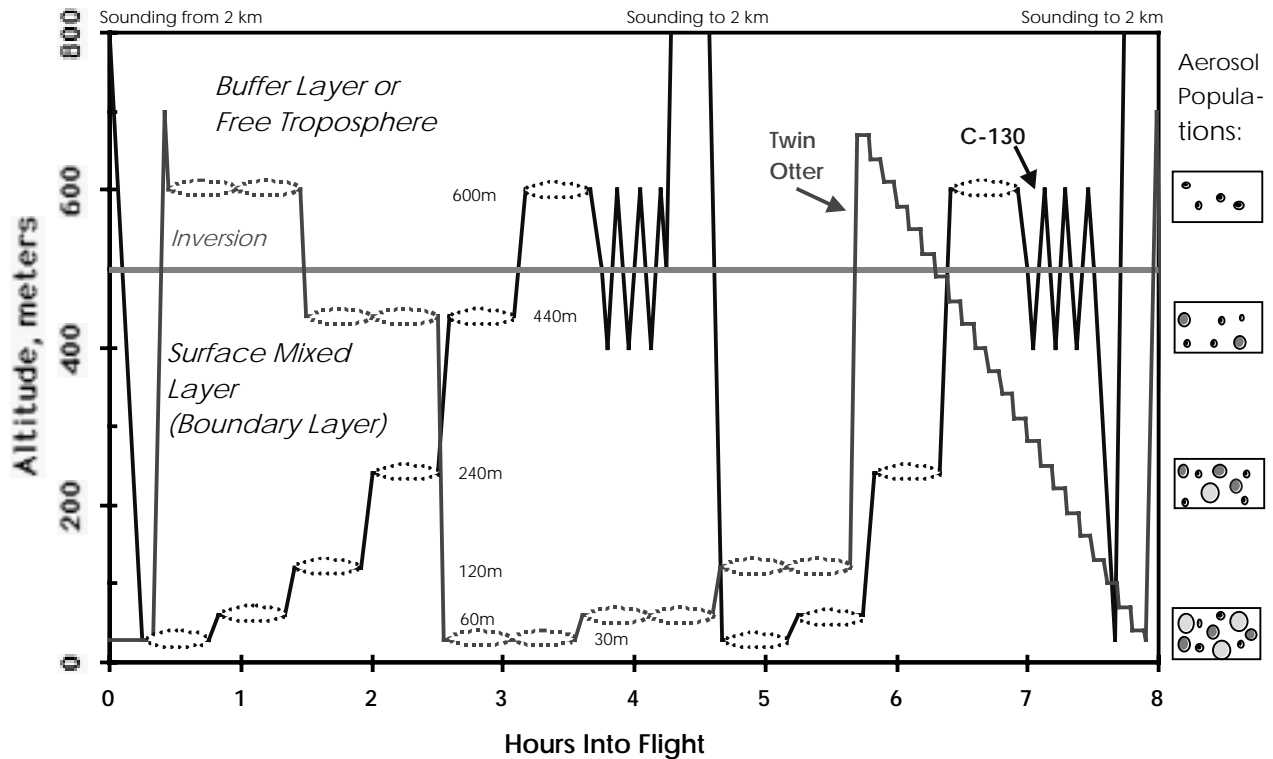


Figure 6. Flight patterns for sea-salt gradient experiment. The C-130 would do 30 minute circles at each of 6 altitudes, repeating four of them, while drifting with the wind. On these legs chemical and physical size distributions would be measured, with an emphasis on differences in the altitude variation of various size classes of seasalt particles. The C-130 would also porpoise through the inversion, to establish the uniformity of its altitude around the circle and to characterize the entrainment rate of BuL or FT air into the BL. The Twin Otter would do 60 minute legs (two 30 min circuits, with a smaller radius than the C-130) at 5 altitudes, to collect a high-volume Moudi sample at each altitude. It would then do a high-resolution gradient sounding, with 5 minutes at each of 22 altitudes. This would allow nephelometers, sulfur gas instruments, and sizing devices enough time to stabilize at each altitude to get good quality data for this detailed gradient.

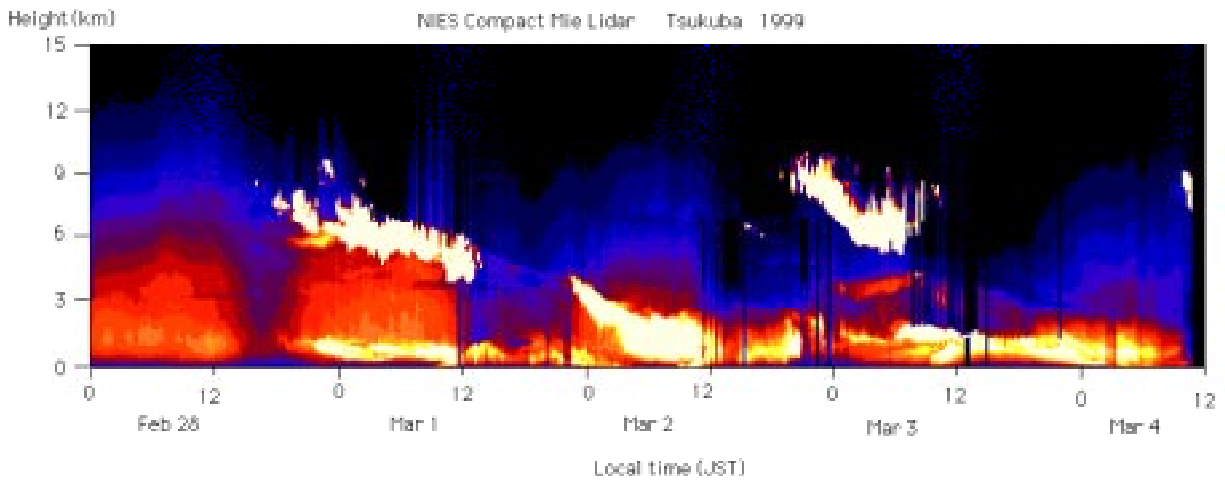


Figure 7. Range corrected Lidar image of a Kosa aerosol layer that appeared at an altitude of approx. 5 km in the evening of March 1st (JST). The layer is clearly seen descending until noon of March 2nd (JST) (N. Sugimoto, I. Matsui). A Similar lidar signal was also recorded at TUMM (T. Murayama). <http://info.nies.go.jp:8094/kosapub/>

ACE-Asia Dust Layers Flight

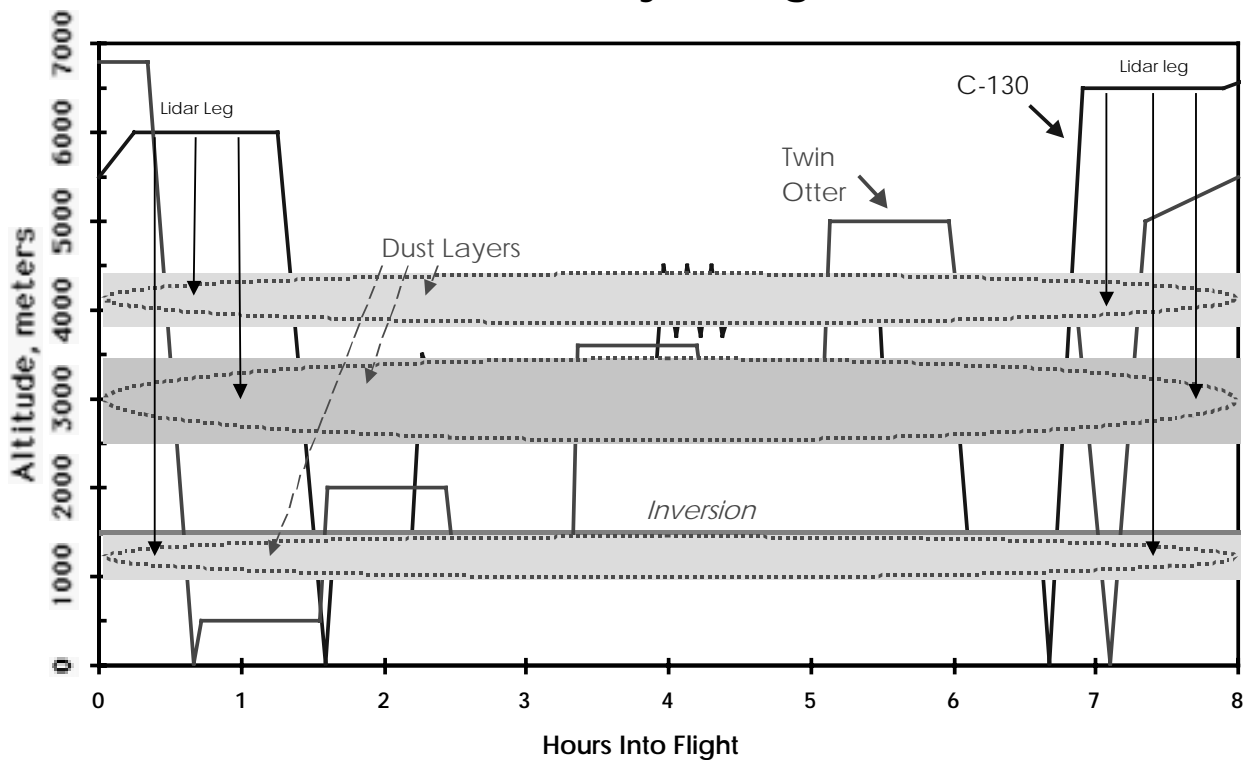


Figure 8. Flight pattern for dust layers study.

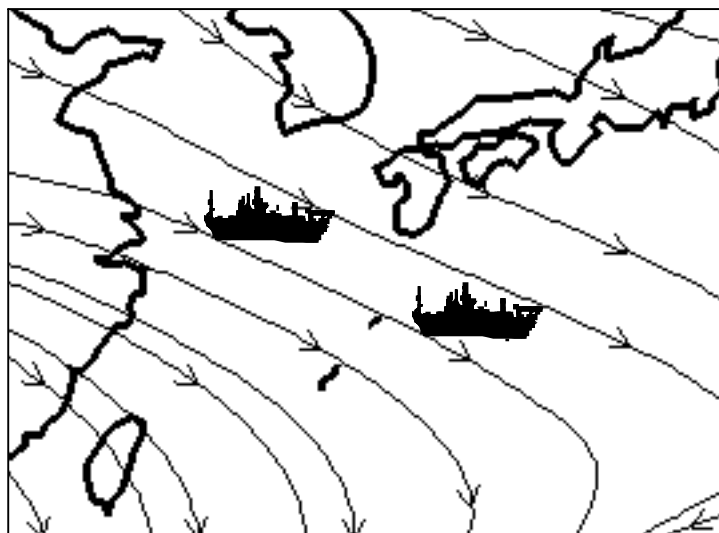


Figure 9. Two ship “Lagrangian” experiment. A balloon or chemical tracer is needed to confirm sampling in the same airmass. Flows by J. Merrill

Stacks of Circular Flight Legs in Lagrangian Experiments

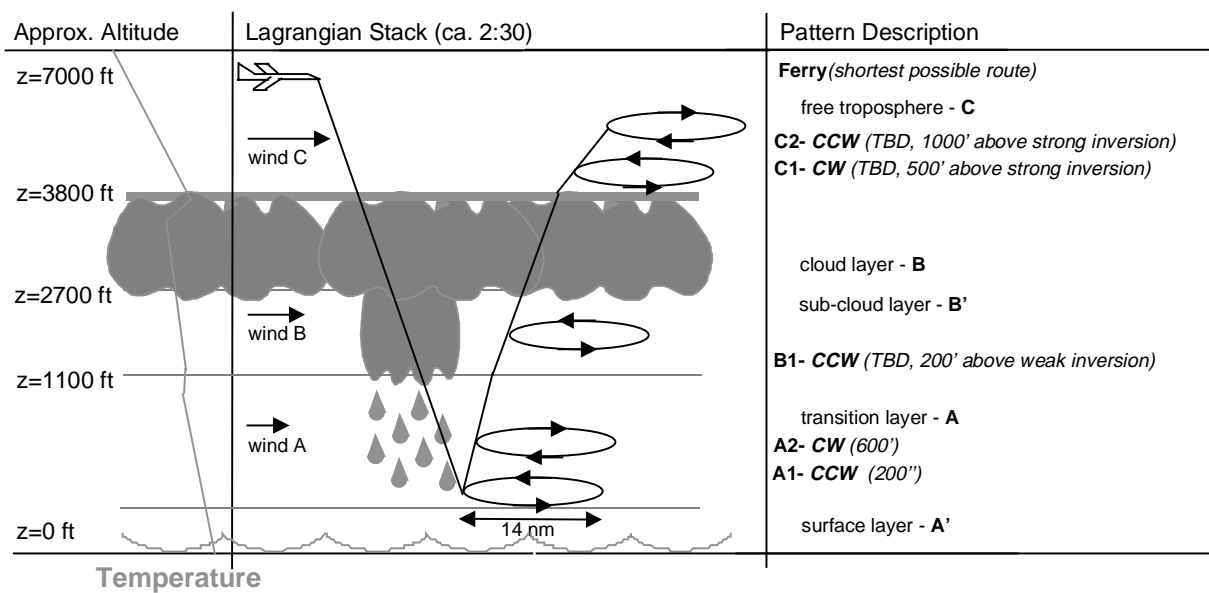


Figure 10. Example of one stack in a Lagrangian flight. L Russell

Two-Aircraft Lagrangian Flights

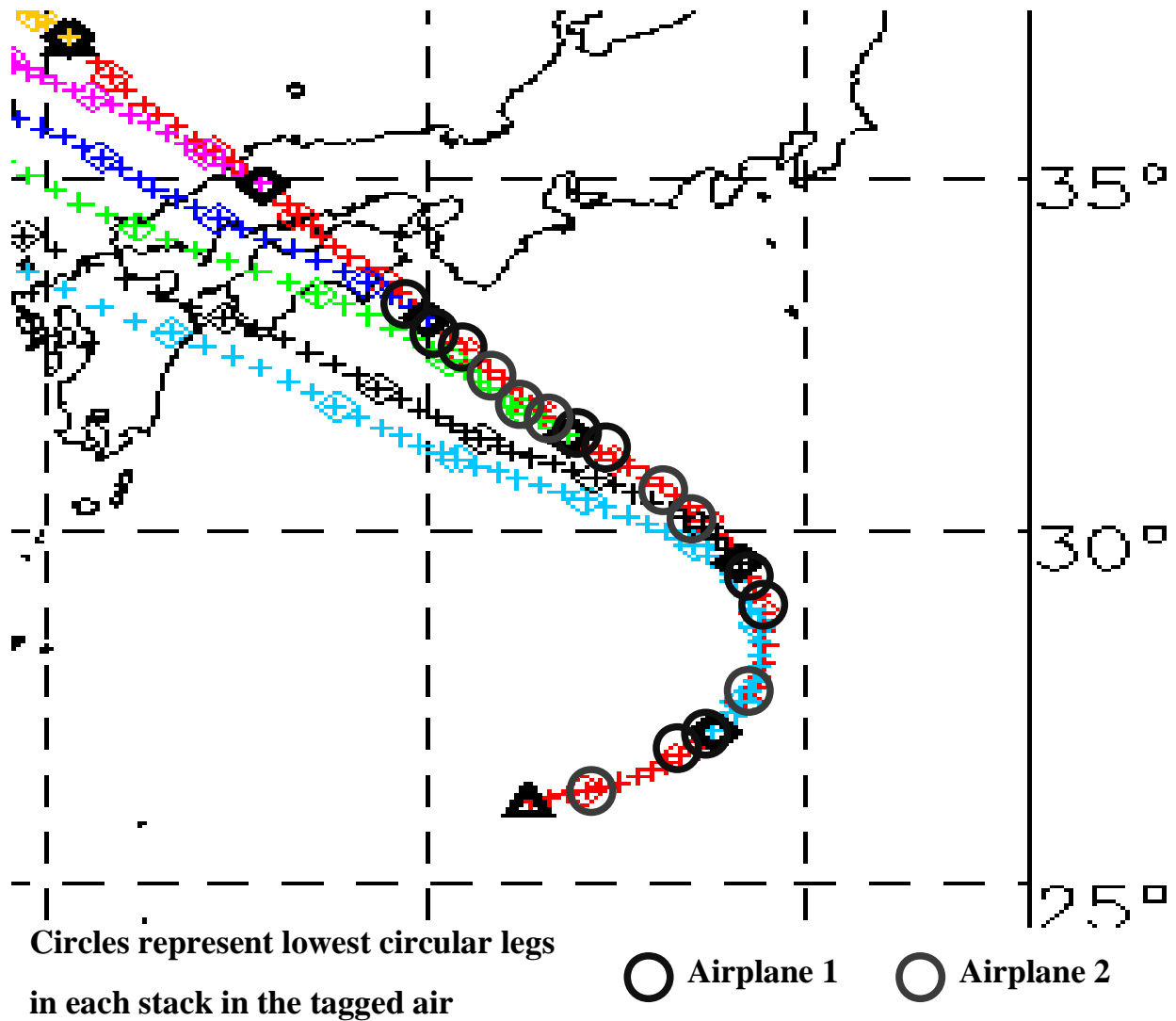


Figure 11. Example of one possible Lagrangian experiment, using two aircraft flying out of Fukuoka, Japan. K. Suhre trajectory

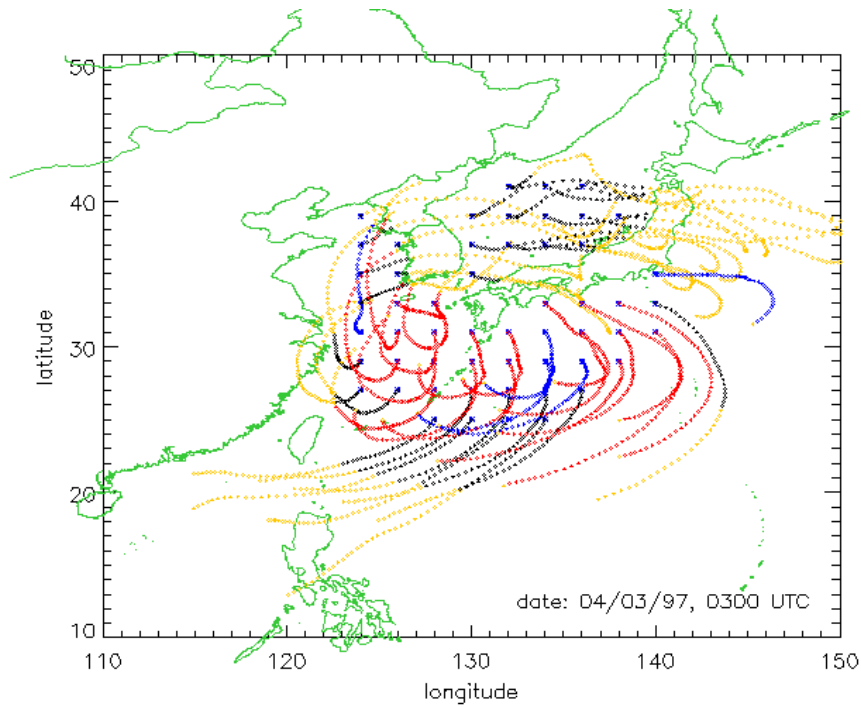
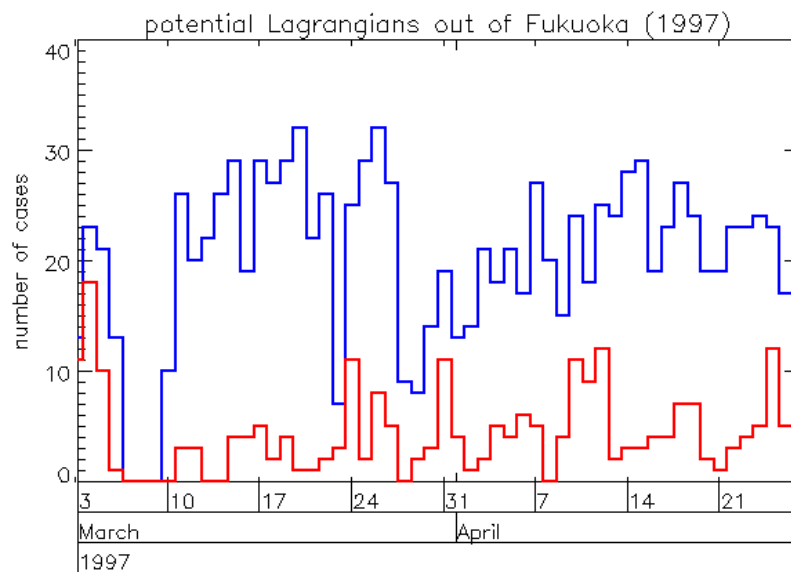


Figure 12. Followable trajectories (in red) on 3 April, 1997. K. Suhre.
<http://www.aero.obs-mip.fr/ace/ace3/trajs/ver2.html>



blue: followable trajectories, red: potential Lagrangian (out of 54)

total: 3024, followable: 1122, pot. Lagrangian: 240

Figure 13. Statistics for “followable” Lagrangian experiments in 1999. K. Suhre.
<http://www.aero.obs-mip.fr/ace/ace3/trajs/ver2.html>

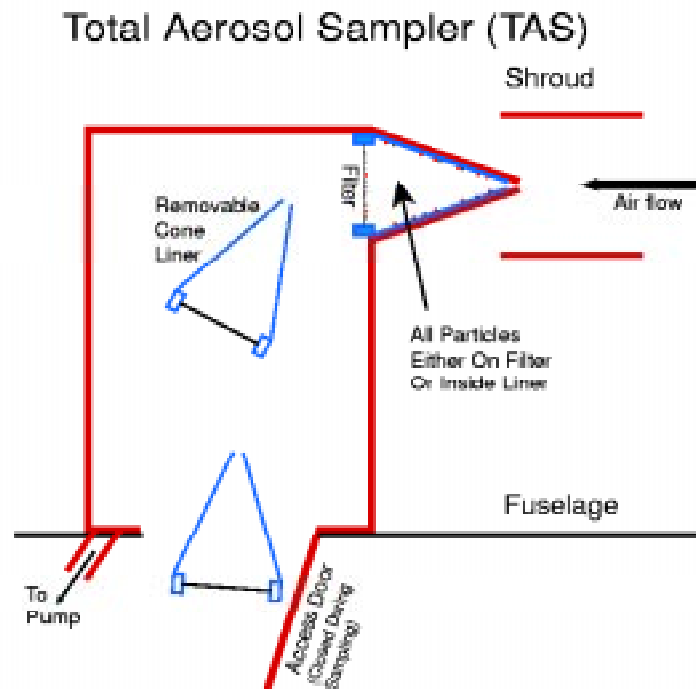


Figure 14. Total aerosol sampler for aircraft. B. Huebert

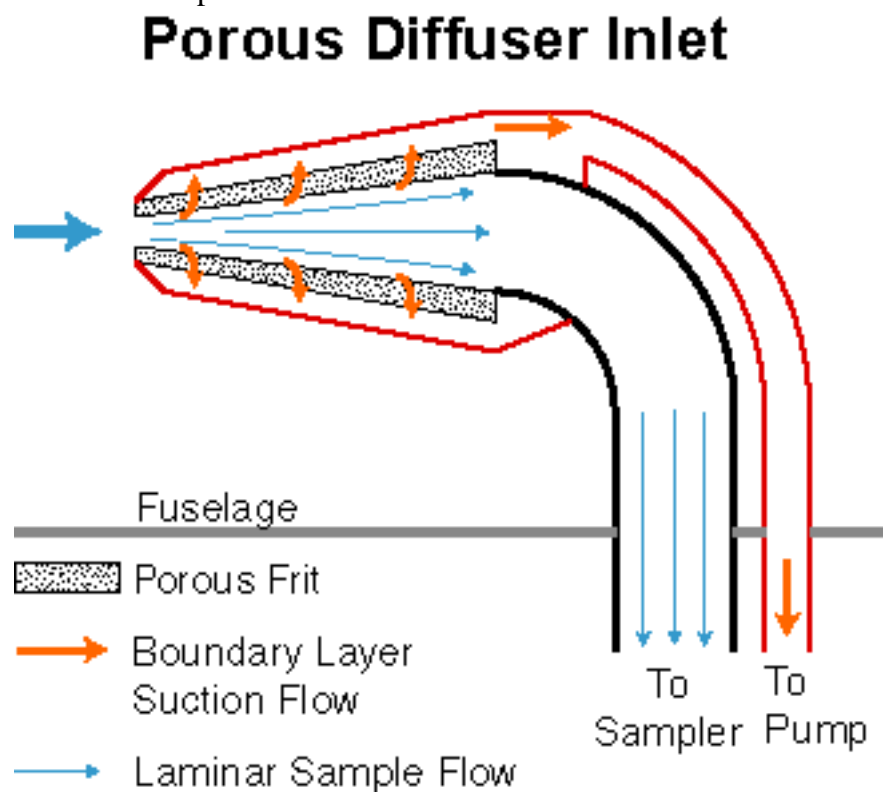


Figure 15. Porous Diffuser Laminar Flow Inlet. By removing boundary layer flow through the walls before it can detach and create turbulence, laminar flow is retained in the diffuser cone and particles are not thrown to the walls. B. Huebert

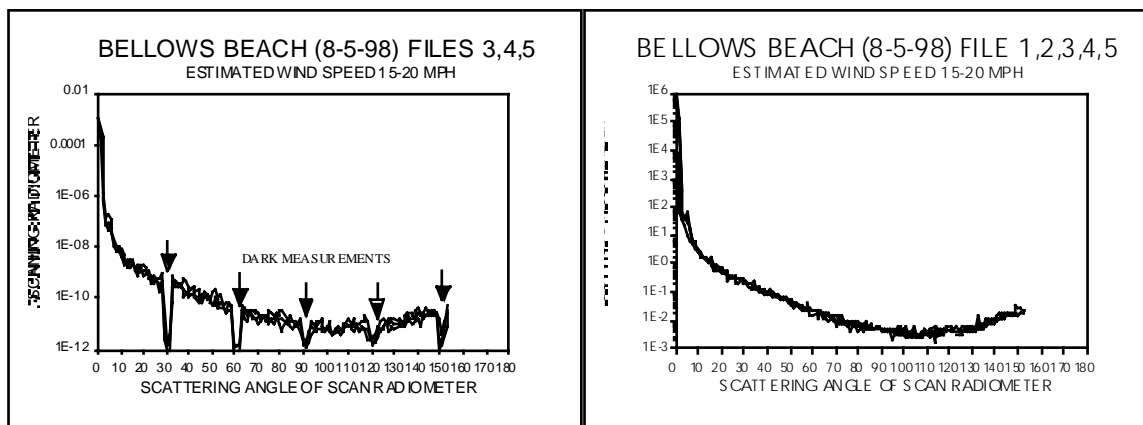


Figure 16. Sea spray aerosol phase function measurement made at approximately 1 degree intervals at 808 nm wavelength. Left panel shows three angular scans with dark measurements. Right panel shows measurements without dark measurements. J. Porter

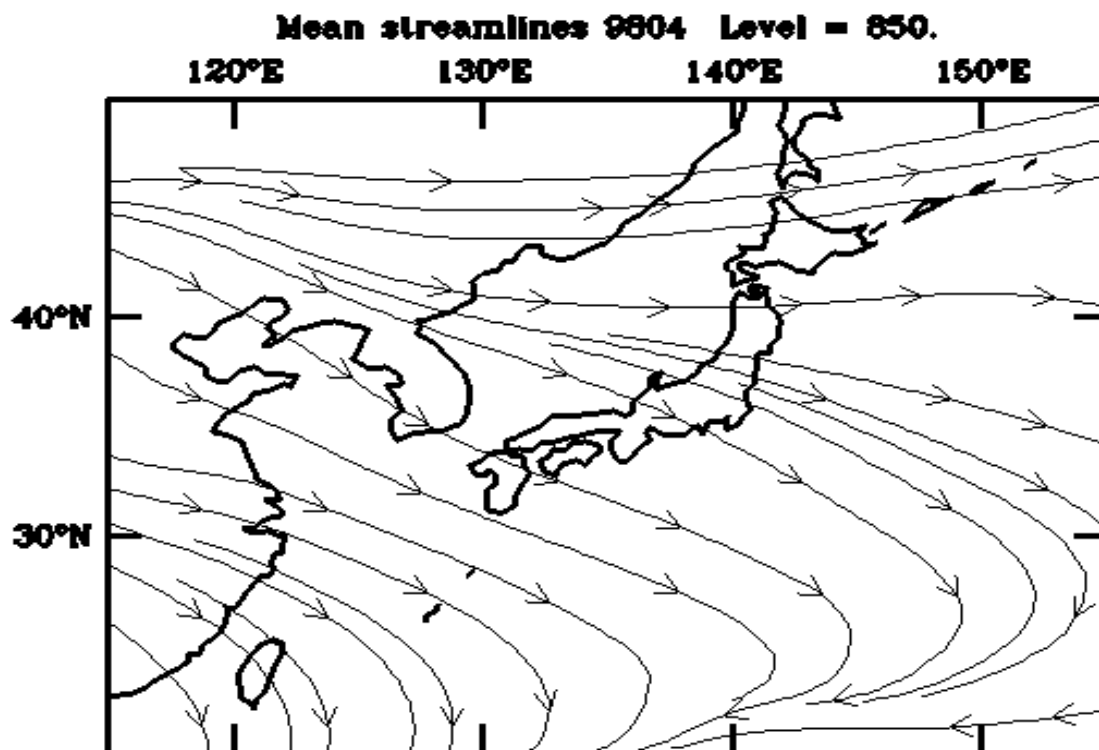


Figure 17. Mean flow for April of 1998 at 850 hPa, approximately 1.5 km above sea level. J. Merrill